Upper Critical Field of the 3 Kelvin Phase in Sr$_2$RuO$_4$

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The inhomogeneous 3 Kelvin phase is most likely a superconducting state nucleating at the interface between micrometer-sized Ru-metal inclusions and Sr$_2$RuO$_4$ above the bulk onset of superconductivity. This filamentary superconducting state yields a characteristic temperature dependence of the upper critical field which is sublinear, i.e., $H_{c2}(T) \propto (T^* - T)^\gamma$ with $0.5 \leq \gamma < 1$ ($T^*$: nucleation temperature). The Ginzburg-Landau theory is used to analyze the behavior of the nucleated spin-triplet phase in a field and the characteristic features of $H_{c2}$ observed in the experiment are explained based on a two-component order parameter in the presence of a filament of enhanced superconductivity with a finite width.

KEYWORDS: Sr$_2$RuO$_4$, upper critical field, filamentary superconductivity, Ginzburg-Landau theory

Sr$_2$RuO$_4$ plays an exemplary role among unconventional superconductors as a realization of spin triplet pairing in a quasi-two-dimensional (2D) Fermi liquid, with some similarities to superfluid $^3$He.1–3 Experiments provide strong evidence of a superconducting state with in-plane equal-spin pairing3 and violation of time reversal symmetry.5 This uniquely identifies the pairing symmetry to be that of a chiral $p$-wave state, analogous to the $A$-phase of $^3$He: $d(k) = \Delta \hat{z}(k_x \pm i k_y)$,6,7 This is a lucky case in many respects. We mention only a few points here. (1) Broken time reversal symmetry is responsible for unusual magnetic properties. (2) The order parameter consists of two complex components $\eta = (\eta_x, \eta_y)$, the only case among all possible triplet pairing states, for tetragonal crystal symmetry, which correspond otherwise to one-component order parameters. Thus, we may write

$$d(k) = \hat{z}(\eta_x k_x + \eta_y k_y).$$

(1)

It gives rise to unusual vortex physics, including a square vortex lattice and anomalous low-temperature flux dynamics.8,9 (4) Chiral gapless subgap quasiparticle states appear at the surface.10,11

For these properties the important feature is the degeneracy of the two order parameter components, which is guaranteed by the tetragonal symmetry. It was suggested that this degeneracy lifted by symmetry lowering states at the interface between the Ru-metal and Sr$_2$RuO$_4$, where the critical temperature is larger possibly due to a locally enhanced density of states and modified electron-electron interactions.15 In such a case the superconducting state appears at a temperature $T^* > T_c$ in a restricted region of lower symmetry. This superconducting state has a single order parameter component and does not violate time reversal symmetry. The component corresponds to the $p$-wave superconducting state with momentum along the interface, i.e., $\eta \cdot n = 0$ where $n$ is the interface normal vector. This filamentary phase yields several unusual properties. Unlike in a conventional $s$-wave superconductor, the transition to the bulk phase is not merely a matter of percolation, but represents a real (time reversal) symmetry-breaking transition. This corresponds to an additional second-order phase transition.16 Moreover, this system may constitute a complex intrinsically phase frustrated superconducting network.

An important way of probing the filamentary phase is the observation of the nucleation in a magnetic field, i.e., the upper critical field $H_{c2}$. The confinement to a narrow filament yields a sublinear temperature dependence $H_{c2}(T) \propto (T^* - T)^\gamma$ where $0.5 \leq \gamma < 1$ in contrast to the linear behavior for the bulk $H_{c2}$.15,17,18 In view of experiments showing exponent $\gamma$ lying between 0.5 and 1 in qualitative agreement with the expectations,19 we would like to reanalyze the behavior of $H_{c2}$ in the filamentary phase.

Our analysis is based on the Ginzburg-Landau model of an infinite planar interface, as introduced in ref. 15. It was suggested that the locally enhanced $T_c$ at the interface is the result of a local lattice distortion mainly by Ru$_4$-octahedra rotation around the $z$-axis. This gives rise to reduced hopping matrix elements such that the Fermi velocities decrease, increasing the density of states with an additional (Stoner) enhancement of the uniform spin fluctuations. The extension $s$ of the distortion is of the order 100 Å and,20 thus, is much shorter than the coherence length $\xi$. On the other hand, the size of the Ru inclusions is $\sim 1 – 10 \ \mu$m, large compared to the coherence length.13,14 Hence we consider here an infinitely extended interface separating two half spaces. The purpose of the present study is to analyze the nucleation...
at the nucleation point. $H_{c2}$ is determined by the instability of $\eta_y$. We use $A = (0, 0, -Hx)$ leading to the equation

$$[-K_2 \partial_x^2 - \xi \sigma \delta(x) + \frac{4K_3 x^2}{l_H^4}] \eta_y = -a \eta_y$$

for $\eta_y$, where $l_H = c\hbar/(eH)$ is the magnetic length. For low fields (long $l_H$), the harmonic potential term is a weak perturbation to (3), so that $\eta_y = \exp(-|x|/\xi_y)$ remains a good approximation. Substituting it into (2) and integrating over $x$ we obtain the second-order term

$$F = a \xi_y + \frac{K_2}{\xi_y} - \xi \sigma + \frac{4K_3 \xi_y^3}{2l_H^4},$$

whose zero determines the instability. Thus $F = 0$ yields $H_{c2}$:

$$H_{c2} = \frac{cH}{2e} \sqrt{\frac{2}{K_3 \xi_y}} \frac{T_y^* - T}{T_c},$$

as found previously. With increasing field, however, $l_H$ becomes shorter and the harmonic potential term in (6) becomes a larger correction. Thus the extension of $\eta_y$ along $n$ shrinks due to the decreasing cyclotron radius. A good approximation to the ground state of the “Schrödinger equation” (6) is obtained by a variational ansatz, which captures the basic behavior in a simple way:

$$\eta_y = \exp(-|x|/\xi_y) \exp(-\sqrt{K_3/K_2} x^2/l_H^4).$$

Here $\exp(-\sqrt{K_3/K_2} x^2/l_H^4)$ describes the asymptotic behavior for distances far from the interface, while the exponential part gives the correct boundary conditions at the interface. Again we substitute $\eta_y$ into (2) and integrate over $x$, so that setting $F = 0$ leads to $H_{c2}(T)$ (Fig. 1). We may approximate the low-field range, by a power law $H_{c2} \propto (T_y^* - T)^\gamma$. We find the best approximation to the variational solution for $\gamma = 0.62$ in the range $0.9T_y^* < T < T_y^* = 2.8$ K. This value compares well with recent experimental findings of $\gamma = 0.7 - 0.75$, which is indeed sublinear.

The exact square-root behavior in the limit of very small fields is experimentally difficult to observe. The limitation of this behavior is given by $\xi \ll l_H$, i.e., $H \ll H^*$ with a characteristic field $H^* = 2\sqrt{K_2/K_3}(\Phi_0/2\pi)/\xi^2 \sim 1T$. Moreover, in Fig. 1 we observe a deviation from our variational solution for $T < 2K$. This is partially due to the limited validity of the Ginzburg-Landau theory which only extends to the region close to $T_y^*$. Furthermore, the suppression of $H_{c2}$ is likely related to a limiting effect (analogous to the paramagnetic limiting) which has also been observed in the bulk $H_{c2}$ for fields in the basal plane. The discussion of this high-field behavior lies beyond our model and our scope.

For $H \parallel n$, we use $A = (0, 0, H y)$ leading to the Ginzburg-Landau equation for $\eta_y$:

$$[-K_2 \partial_y^2 - K_1 \partial_y^2 + \xi \sigma \delta(x) + \frac{4K_3 y^2}{l_H^4}] \eta_y = -a \eta_y.$$
where $C$ and $H^x$ are the characteristic field and magnetic field. We can integrate the free energy analytically to fix the transition temperature $T_x^c$ to be above 1 K.

Fig. 1. Temperature dependence of the upper critical field for an in-plane field. The solid line denotes $H \parallel y$. For a low magnetic field, $H_{c2}$ has a square-root dependence $H_{c2} \propto (T_x^c - T)^{5/2}$, which is plotted as a dashed line for $H \parallel y$. The long-dash linear line denotes $H \parallel x$. We choose the following parameters: $K_5/K_1 = 1/500$ and $K_1/(ch/2c) = \frac{\xi^2}{(ch/2c)} = 20$. $\sigma$ is given to determine the upper critical field explicitly. We plot the result in Fig. 2. Our result well reproduces the experimental result above $0.9T_y^c$. In this region, we find an exponent $\gamma = 0.76$ fit to a power law, which agrees well with the experimental result ($\gamma = 0.72$). In the vicinity of $T_y^c$, $H_{c2}$ exhibits again a very-low-field square-root dependence as in the case of $H \parallel y$, which is plotted in Fig. 2 as a dashed line. In this case, $H_{c2}$ deviates even more rapidly from the square-root behavior as temperature decreases than the in-plane fields. The reason lies in the lower characteristic field $H^* = 2\sqrt{K_x/K_1}/(2\pi)\xi^2 \sim 0.05T$.

Fig. 2. Temperature dependence of the upper critical field for $H \parallel z$. For quite low magnetic fields, $H_{c2}$ has a square-root dependence, which is plotted as a dashed line. The parameters are the same as in Fig. 1. Circles denote the experimental data.

Since the $x$- and $y$-dependences factorize, we obtain the relevant solution:

$$\eta_y = \exp(-x/\xi_y) \exp(-\sqrt{K_5/K_1}y^2/\xi_H^2),$$

and $H_{c2} \propto T_y^c - T$ as shown in Fig. 1 (long-dash line). The nucleation field for this direction is obviously lower. It is clear that the observed $H_{c2}$ is due to interfaces that lie parallel to the applied field ($H \perp n$), which yields the highest nucleation field.

Now we turn to fields parallel to the $z$-axis assuming simultaneously $H \perp n$. In this case the two order parameter components are coupled. We choose the vector potential as $A = (0, Hx, 0)$ so that the free energy is expressed as

$$F = \int_0^\infty dx (f_x + f_y + f_{xy}),$$
$$f_x = \frac{d|\eta_x(x)|^2 + K_1(2)|\partial_x \eta_x(x)|^2}{4K_2(1)x^2} \left| \frac{\eta_x(x)}{\xi_H} \right|^2,$$
$$f_y = 2(\frac{K_3 + K_4}{\xi_H^2}) \left( \eta_x \partial_x \eta_y \eta_y \partial_x \eta_y + \eta_y \partial_x \eta_x \eta_x + \text{c.c.} \right).$$

As in the case of $H \parallel y$, we introduce a variational form for the order parameters:

$$\eta_{y(x)} = C_{y(x)} \exp\left(-\frac{|x|}{\xi_{y(x)}}\right) \exp\left(-\sqrt{\frac{K_1(2)}{K_2(1)}x^2}\right),$$

where $C_{y(x)}$ are coefficients to be determined in order to maximize the nucleation temperature for a given magnetic field. We can integrate the free energy analytically and determine the upper critical field explicitly. We plot the result in Fig. 2. Our result well reproduces the experimental data.

In the vicinity of $T_y^c$, $H_{c2}$ exhibits again a very-low-field square-root dependence as in the case of $H \parallel y$, which is plotted in Fig. 2 as a dashed line. In this case, $H_{c2}$ deviates even more rapidly from the square-root behavior as temperature decreases than the in-plane fields. The reason lies in the lower characteristic field $H^* = 2\sqrt{K_x/K_1}/(2\pi)\xi^2 \sim 0.05T$.

In Fig. 2 the experimental data show a pronounced upturn for $T < 2K$, opposite to the trend for in-plane fields. We would now like to discuss the origin of this behavior by extending our model. So far we have assumed that the extension of the region with enhanced superconductivity is negligible and is well described by a delta function. We replace, however, now the delta function in eq.2 by

$$\delta(x) \rightarrow \frac{1}{\sqrt{\pi s}} \exp\left[-\frac{(x/s)^2}{2}\right],$$

where $s$ represents the width of the interface region of enhanced transition temperature. In the introduction we speculated that this region is characterized by an increased density of states or, equivalently, by a decreased Fermi velocity. Since the coefficient of $K_i$ ($i = 1, 2, 3$ and 4) is connected with the Fermi velocity in the $x-y$ plane ($K_i \propto v_F^2/T^2_c$), we introduce in addition, the following spatial dependence in $K_i$:

$$K_i \rightarrow K_i(1 - \lambda \exp[-(x/s)^2]).$$

with $\lambda (0 < \lambda < 1)$ as a parameter. Assuming the same variational order parameter form we integrate the free energy analytically. The resulting $H_{c2}$ is shown in Fig. 3. In particular, we observe the onset of an upturn of $H_{c2}$ for $H \parallel z$ at approximately 2 K, which compares well with the experiment.

This feature originates from the fact that with shrinking magnetic length the region of nucleation becomes...
increasingly confined into a narrower layer, where we find an enhanced transition temperature as well as a shorter local coherence length. Both act to increase the critical field. Thus the onset of the upturn is expected when $l_H \sim s$. The fit to the data with our variational approach yields $s \approx 200 \text{Å}$. Note that this kind of upturn behavior is not expected for in-plane fields, since $K_4$, that determines the coupling, would not have the form (15) as it depends on the $z$-axis Fermi velocity that would not be significantly affected by RuO$_4$-octahedra rotations.

In summary, the discussion of the upper critical field for the filamentary superconducting phase exhibits several length scales to be taken into account, which are the effective magnetic length $l_H$, the coherence length $\xi$, and the extension $s$ of the interface regions. If the effective magnetic length $l_H$ is much larger than $\xi$ and $s$, we find that $H_{c2}$ would follow the square-root power law $(T_g - T)^{1/2}$. Once the field starts to shrink the extension of the nucleated order parameter, we encounter a deviation from this behavior and a power law fit would yield a different power law. Our discussion shows that in the case of the 3-Kelvin phase the square-root behavior occurs in a very limited range of low fields which is hard to analyze. Finally, if the field becomes sufficiently strong to confine the nucleating order parameter in the interface region $s$ a relative increase of the upper critical field is possible. In the 3-Kelvin phase this is observed for the field along the $z$-axis. However, it has to be noticed that an additional important enhancement factor is the coupling of the two order parameter components. This latter effect is due to the Zeeman coupling of the magnetic field to the orbital magnetic moment of the Cooper pair for the chiral $p$-wave phase.

The comparison of our theory with the experiment shows that we are in principle able to fit the experimental data. However, this aspect has to be viewed with care, as the Ginzburg-Landau free energy is expanded at a temperature around $T^*$ and has therefore limited quantitative reliability. Moreover the model was in many respects simplified. Nevertheless, the ability to reproduce the qualitative features are convincing, we believe. One obvious problem is the limiting behavior for in plane fields which seems to be present in both the bulk and 3 Kelvin phase of Sr$_3$RuO$_4$.

The upper critical field may be considered a strong confirmation of the assumption that the 3 K phase Sr$_2$RuO$_4$ is due to the local enhancement of the superconducting transition temperature at the interface of Ru-inclusions and Sr$_3$RuO$_4$. Thus, we expect that Sr$_2$RuO$_4$ has two consecutive phase transitions due to the symmetry lowering by the Ru inclusions. This fact still remains to be experimentally verified. It could then be added to the other convincing evidence for chiral $p$-wave pairing in Sr$_2$RuO$_4$.

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