Quantum Criticality Beneath the Superconducting Dome in $\beta$-YbAlB$_4$

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Abstract. Yb-based heavy fermion superconductor $\beta$-YbAlB$_4$ at 0 K and 0 T at ambient pressure is located near the quantum critical point with strong mixed valiancy. In this type of Yb electron system, we expect that the magnetic order connected to the quantum critical point derives from the applied pressure. We built a pressure–temperature phase diagram for $\beta$-YbAlB$_4$ by measuring the electrical resistivity of high quality single crystal at temperatures down to 40 mK under an applied pressure. A strange metal region appeared, showing non-Fermi liquid $\rho_{ab} \propto T^{1.5}$ behavior, which is stable with applied pressure up to 0.4 GPa, even when below the superconducting dome excluded by a magnetic field of 0.1 T. By increasing of pressure above 2.5 GPa, a magnetic order is first generated. Such ambient quantum criticality/superconductivity is unconventional and is detached from the magnetic order.

1. Introduction
Materials near the quantum critical point (QCP) have attracted much attention because they exhibit unconventional superconductivity and exotic strange metal behavior. In particular, high-temperature superconductivity in Cu-based and Fe-based materials near the QCP and unconventional metal behavior in heavy fermion (HF) materials near the QCP are well known phenomena. In 4$f$-electron HF systems, the QCP appearing at 0 K between distinct ordered and disordered quantum phases, is intrinsically unstable and exhibits unconventional metal behavior, becoming a non-Fermi liquid dominated by quantum fluctuations rather than thermal fluctuations. The antiferromagnetic quantum fluctuations near the QCP also are believed to produce unconventional superconductivity, which is indeed connected to the presence of a magnetically ordered phase and is mediated by magnetism. [1, 2, 3] Conversely, these superconducting states emerge to stabilize the destabilized region near the QCP. The non-Fermi liquid state—also known the strange metal near the QCP, and which sometimes appears with unconventional superconductivity—shows characteristic power-law temperature-dependent physical properties such as electrical resistivity $\rho(T)$, specific heat $C(T)/T$, and magnetization $m(T)$ depend anomalously on temperature. Moreover, the quantum phase transitions across the QCP can be accessed by varying a physical parameter—such as magnetic field, external pressure, or chemical composition. Thus, by slightly tuning a physical parameter in an HF material at its QCP, the material easily transitions to a stabilized phase, such as a magnetically ordered phase or Fermi liquid phase. This quantum phase transition and its criticality have been studied and is well described by the conventional Doniach phase, mostly in the Ce-based HF systems.
Recently, however, exotic QCP behaviors have been reported in Yb-based HF system, which is interesting because of the different electron-hole analogy of $4f^{13}$-Ce$^{3+}$ and $4f^{11}$-Yb$^{3+}$. Yb-based HF systems are model materials with strong mixed valency for studying quantum criticality, which is not found in Ce-based HF systems.

The prototypical Yb-based HF system YbRh$_2$Si$_2$ is a weak antiferromagnet below $T_N = 0.07$ K with the intermediate valence state of Yb ($Yb^{2.9+}$) and can transform into an unconventional non-Fermi liquid when doped with 5% Ge in its Si site. To investigate the behavior of YbRh$_2$Si$_2$ near the QCP, they substituted Rh atoms with $A$ atoms in Yb(Rh$_{1-x}A_x$)$_2$Si$_2$ as a form of chemical pressure, substituting Co and Ir for $A$ as positive and negative chemical pressure, respectively. At the QCP, YbRh$_2$Si$_2$ shows an antiferromagnetic transition temperature with Kondo breakdown. Conversely, Co-doped YbRh$_2$Si$_2$ shows the signature of Kondo breakdown within the magnetically ordered phase. For Ir-doped YbRh$_2$Si$_2$, the Fermi liquid state is not connected with the antiferromagnetic order at a single QCP. These phase diagrams cannot be described by a Doniach phase, so the Yb-based HF system promises to display new behavior near the QCP. [6]

In contrast, $\beta$-YbAlB$_4$ shows a QCP at 0 K without tuning any parameter such as field or composition. [7] This material also exhibits a strong intermediate valence state ($Yb^{2.73+}$ at 20 K). [8] Moreover, its material is the first superconductor $T_c = 85$ mK) near a QCP observed in the Yb-based HF family. The existence of $T_c$ in HF compounds is also an indicator of how far the current state is from the QCP. The fact that a superconducting state exists in ambient $\beta$-YbAlB$_4$ indicates that it is very near to quantum criticality. Indeed, its anomalous paramagnetic properties suggest that NFL exists in transport ($\rho_{ab} \propto T^{3/2}$), magnetization ($\chi_c \propto T^{-1/2}$), and specific heat ($C_M/T \propto -\ln T$) at low temperatures. [9, 7] The $T_c$ is also very sensitive to reductions in the residual resistivity ratio (RRR): $T_c$ decreases with increasing RRR and also disappears near 1% chemical Fe doping in the Al site. The low degree of impurity concentration or lattice deformation likely destroys the superconducting state in the pure Kondo lattice system and presumably affects the quantum critical behavior at very low temperatures. Thus, in studying the QCP it is important to tune its state with maintaining material quality, which is only possible in exactly the same high-quality single crystal.

Applying pressure is a non-thermal way to tune the QCP, and applying physical pressure is better than applying chemical pressure from changing the composition because chemical doping induces local lattice disorders or impurity effects. Here, to tune the ground state without causing lattice disorder, we pressurized an extremely high-quality single crystal of $\beta$-YbAlB$_4$ and investigated its QCP beneath the superconductivity dome.

2. Experimental details

Single crystals of $\beta$-YbAlB$_4$ were grown by the aluminum self-flux method. [10] To test their crystalline quality, we measured their residual resistivity ratio $\text{RRR} = \rho_{ab}(300 \text{ K})/\rho_{ab}(0 \text{ K})$. By testing 20 to 30 crystals, we obtained a few single crystals with an RRR greater than 250. We spot-welded electrical contacts to the surface of the crystals using 20 $\mu$m Au wires. The in-plane AC electrical resistivity $\rho_{ab}$ was measured down to 40 mK by using a $^3$He/$^4$He dilution refrigerator equipped with low-temperature transformers, which provide amplification of a factor of 30. Pressure was applied using a hybrid CuBe/NiCrAl piston-cylinder-type cell with a transmission medium of Daphne 7373. [11] No hysteresis in resistivity appeared between warming and cooling runs. The thermal gradient across the pressure cell, monitored by two thermometers (RuO$_2$ tips) at the top and bottom of the cell, was less than 5 mK. The superconducting transition temperatures, Al ($T_c = 1.19$ K) and Pd ($T_c = 3.9$ K), were used as accurate measures of pressure at low temperature. [12, 13]
3. Results and Discussion
To investigate the quantum criticality of Yb-based heavy fermion materials, we probed an ultrapure single crystal (RRR=300) of $\beta$-YbAlB$_4$ and performed highly sensitive transport measurements at low temperature. We measured these transport properties under pressures as high as 3 GPa and at temperatures of 0.04–1.5 K. Figure 1(a) shows the temperature-dependent electrical resistivity $\rho_{ab}(T)$ under zero field parallel to the $ab$-plane. Over a wide temperature range, $\rho_{ab}(T) \sim T^{1.5}$ appeared under a pressure of 0.25 GPa. Moreover, the applied pressure shifted $\rho_{ab}(T)$ by a constant resistivity, but the trends in resistivity remained the same. [14, 15] This result shows that the pronounced NFL behavior of resistivity did not change under an applied pressure of 0.25 GPa, although the pressure did decrease $T_c$.

![Figure 1](image-url)

**Figure 1.** Zero-field resistivity data under various pressures, nearly overlapping but shifted by a constant resistivity indicated in the parentheses. (b) Resistivity data taken under 0.1 T, nearly overlapping but shifted by a constant resistivity indicated in the parentheses.

$\beta$-YbAlB$_4$ is reported to be the anisotropic type-II superconductor. This critical field $B_{c2}$ shows an anisotropic upper critical field of $B_{c2} \sim 300$ G along $B//c$, which is lower than $B_{c2} \sim 1500$ G along the $ab$-plane. A magnetic field of 0.1 T restores the anomalous normal state, reducing $T_c$ to 40 mK. [16] In contrast, applying a magnetic field parallel to $ab$-plane shifts the material into the Fermi liquid region. However, the resistivity was not sensitive to the field along the $ab$-plane, as shown in our previous study. In an applied field along $ab$-plane, the non-Fermi liquid state hidden below $T_c$ emerges. Experimentally, the power law $\alpha$ determined from $\rho_{ab}(T)$ under a field of 0.1 T shows no pressure dependence, as shown in Figure 1(b). [14]

To evaluate the quantum criticality near 0 K in this material, $T_c$ is an unwelcome complication. Figure 2 shows a contour plot giving the resistivity power-law exponent versus pressure. At a low pressure of $P \leq 0.72$ GPa, the resistivity data under 0.1 T were used to exclude $T_c$. The original data under an ambient field was measured under high pressures of $0.72 \leq P \leq 2.7$ GPa. The temperature dependence of the power-law exponent is determined by $\alpha = \partial \ln(\rho_{ab}(T) - \rho_0)/\partial T$. [14, 15] The Fermi-liquid temperature $T_{FL}$ was determined as $\alpha = 2\pm0.1$. From the phase diagram, we obtained the following results: (1) At high pressure, a strange metal region appeared, the so-called “quantum-critical like phase,” beneath superconducting dome, although this material is known to exhibit intrinsic zero-field quantum criticality at ambient pressure. [7] (2) A wide range of quantum criticality hidden by the superconducting phase is isolated from the border of magnetism. (3) Neither the quantum critical-like phase nor the isolated superconducting phase are predicted by the standard theory based on quantum spin fluctuations. A similar quantum-critical-like phase separated from the
Figure 2. Contour plot of the resistivity power-law exponent $\alpha$ as a function of temperature and pressure. At the low pressure of $P \leq 0.72$ GPa, the data taken under 0.1 T ($B//ab$-plane) were used to extrapolate the analysis down to the lowest temperature by suppressing the superconductivity. $T_{\text{FL}}$ is defined as the temperature where the exponent reaches 2. The values of $T_{\text{FL}}$ under zero field and 0.1 T are given by blue and purple circles, respectively. The solid blue line is a guide for the eye. The vertical solid black line indicates $P = 0.72$ GPa.

conventional antiferromagnetic QCP appears in Ir-doped YbRh$_2$Si$_2$ in an applied magnetic field. [6] At 6% Ir, the phase reveals a strong divergence of $C/T$, presumably showing a new type of metallic spin-liquid ground state. [17] The same type of mechanism presumably arise, for instance, by frustrating the Yb honeycomb lattice in $\beta$-YbAlB$_4$. The extended Doniach phase diagram—the QK phase diagram of the Kondo screening (K) and spin zero-point fluctuations (Q)—suggested by recent theoretical approaches would likely explain a number of experimental systems, including YbAgGe and doped YbRh$_2$Si$_2$, and even $\beta$-YbAlB$_4$. [4] Another possible reason for the stabilized phase may be the valence fluctuations of Yb ions. [8] In $\beta$-YbAlB$_4$ we confirmed the possibility of a quantum-critical-like phase by studying a very pure sample with a pure Kondo lattice under zero field.

4. Summary
This study of $\beta$-YbAlB$_4$ suggests that a new unstable region exists, a non-Fermi liquid state, between the stabilized phases, that is not similar to a single critical point in conventional critical phenomena. This finding appears to be the first evidence of a quantum phase hidden by the superconducting dome in HF materials. The Fermi liquid behavior appeared over a wide pressure range ($0.4 \leq P \leq 2.5$ GPa) between the non-Fermi liquid region and the magnetically stable region, but under ambient pressure the material exhibited a real QCP, separated from 2nd QCP near 2.5 GPa. Here, the $T_c$ was as low as 85 mK near the quantum critical point. Thus, the new region below the superconducting dome could appear in a small field. This type of study would be impossible for high-$T_c$ superconductors, such as Cu-based and Fe-based high-
$T_c$ superconductors, because an extremely high magnetic field is needed to exclude the high temperature superconducting state and will destroy the ground state.

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