Anomalous low-field diamagnetic response in ultraclean URu_2Si_2 superconductor

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Abstract. The lower critical field \(H_{c1}\) in ultraclean URu_2Si_2 single crystals \((T_c = 1.4\,\text{K})\) with residual resistivity ratio exceeding 700 is precisely determined by the positional dependence of the local magnetic induction measured down to 55 mK. We show that the whole \(H_{c1}(T)\) for \(H\parallel a\) can be explained by the two gaps with line and point nodes. In contrast, for \(H\parallel c\) we find a distinct kink in the slope of \(H_{c1}\) at 1.2 K, which cannot be accounted for by the two gaps. This outstanding anomaly demonstrates a highly unusual superconducting state embedded in the hidden order state of URu_2Si_2 possibly related to the time reversal symmetry breaking.

1. Introduction

The heavy-fermion compound URu_2Si_2 has mystified researchers since the superconductivity occurs deep inside the mysterious ‘hidden order’ state (whose transition temperature is \(T_h = 17.5\,\text{K}\)) [1, 2]. Below \(T_h\), most of the carriers (>90%) disappears owing to the formation of the electronic excitation gap at a large portion of the Fermi surface, resulting in a semimetallic electronic structure, and the remaining small number of carriers undergo a transition into the exotic superconducting state below \(T_c = 1.4\,\text{K}\) [3].

Here, to shed further light on the unusual superconducting properties of URu_2Si_2, we measured low-field diamagnetic response of ultraclean single crystals by using miniature Hall sensors, from which the lower critical field \(H_{c1}\) is accurately determined down to very low temperatures [4]. We find an anomalous kink structure in the temperature dependence of \(H_{c1}\) for \(H\parallel c\), which has not been reported in the samples with low residual resistivity ratio \((RRR)\) [5]. This anomaly suggests that the superconducting state of ultraclean URu_2Si_2 is highly unusual, and possible implications of these findings are discussed.
2. Experimental
Experiments have been performed on URu$_2$Si$_2$ single crystals (with typical dimensions of $2.3 \times 0.75 \times 0.15$ mm$^3$) with very large RRR value exceeding 700 [6]. Reliable determination of $H_{c1}$ is a difficult task in the presence of the flux pinning and magnetic relaxation. To avoid this difficulty, we use a method by determining $H_{c1}$ as the field $H_p$ at which first flux penetration occurs from the edge of the crystal [7]. For this purpose, we used a miniature Hall-sensor array tailored in a GaAs/AlGaAs heterostructure with each active area of $\sim 5 \times 5$ $\mu$m$^2$ (the center-to-center distance of neighboring sensors is 20 $\mu$m) [8]. The crystal is directly placed on top of the array, as illustrated in the inset of Fig. 1(c).

3. Results and discussion
Figures 1(a) and (b) display the field dependence of local induction $B_{\text{edge}}$ measured by the Hall sensor near the edge for $H \parallel a$ and $H \parallel c$, respectively. The flux penetration fields $H_p$ shown by the arrows are clearly resolved by the deviation from the Meissner state ($B_{\text{edge}} = 0$). Figure 1(c) displays the positional dependence of $H_p$. $H_p$ increases with increasing the distance $d$ from the edge at low temperatures. However, $H_p$ is position-independent close to the edge ($d \leq 40$ $\mu$m) even at low temperatures. These results demonstrate that we can accurately determine the lower critical field from $H_p$ measured by sensors near the edge.

Figures 2 depict the temperature dependence of $H_{c1}^a$ and $H_{c1}^c$, where $H_{c1}^a$ and $H_{c1}^c$ are the lower critical field for $H \parallel a$ and $H \parallel c$, respectively. The absolute value of $H_{c1}$ is obtained by taking into account the demagnetization effect. For a platelet sample, $H_{c1}$ is given by $H_{c1} = H_p/\tanh(0.36b/a)$, where $a$ and $b$ are the width and the thickness of the crystal, respectively [9]. In the present crystal, $H_{c1}$ are evaluated as $H_{c1}^a = 3.82H_p$ and $H_{c1}^c = 1.15H_p$. As seen in Figs. 2, $H_{c1}^c(T)$ exhibits a distinct change in the slope with a kink at $T_0 \simeq 1.2$ K and $H_{c1}^a(T)$ exhibits a cusp behavior at $T_p \simeq 1.0$ K. At $T_p$ the specific heat measurements have reported no anomalies in the crystals with $RRR > 100$. However, a peak structure in the specific

![Figure 1](image1.png)

**Figure 1.** (a) Local magnetic induction $B_{\text{edge}}$ measured by the sensor at the edge of the crystal as a function of $H$ for $H \parallel a$. The arrows indicate the flux penetration field $H_p$. Each data is vertically shifted for clarity. (b) The same plot for $H \parallel c$. (c) Positional dependence of $H_p$. Inset: Schematic illustration of the experimental setup. Sensor 1 locates just outside of the crystal.
Figure 2. Temperature dependence of the lower critical fields $H_{c1}$ for (a) $H \parallel a$ and (b) $H \parallel c$ (open symbols). Arrows indicate a kink anomaly at $T_Q = 1.2$ K and a cusp behavior at $T_p \approx 1$ K. Normalized superfluid densities (a) $\sqrt{n_s^a n_s^c}$ and (b) $n_s^a$ are also plotted. The dotted and dashed lines indicate the superfluid densities for the hole band with $\Delta_h = 1.6 k_B T_c$ and the electron band with $\Delta_e = 4.0 k_B T_c$, respectively, which are obtained by assuming the illustrated nodal topologies suggested by the thermal conductivity measurements [3]. The solid thick and thin lines represent the results of the two-gap fitting and the calculation for $s$-wave gap, respectively.

Heat has been reported in the vicinity of $T_p$ for crystals with low RRR values, possibly due to the inhomogeneous distribution of $T_c$ [6]. Therefore we cannot rule out a possibility that the anomaly at $T_p$ in $H_{c1}^a$ is due to a tiny portion with low-$T_c$ phase in the crystal, though no specific heat anomaly is observed in the present crystal at $T_p$. As for the kink at $T_Q$, there is no evidence for the low-$T_c$ phase at this temperature. So we infer that the pronounced kink in the slope of $H_{c1}^c$ found in the ultraclean crystals is intrinsic in the superconducting state of URu$_2$Si$_2$.

We next discuss the temperature dependence of $H_{c1}$ in terms of the two-gap model since it has been shown below $T_Q$ the nearly perfect compensation (equal number of electrons and holes) is realized in URu$_2$Si$_2$ [3]. In the two-gap model, the in-plane and out-of-plane superfluid density normalized by their values at $T = 0$ K, $n_s^a$ and $n_s^c$, respectively, can be written as $n_s^i(T) = x^i n_s^i(0) + (1 - x^i) n_s^i(T)$ where $n_s^i$ and $n_s^j$ are the normalized superfluid density of hole and electron bands, respectively, and $x^i$ is the ratio of the electron and hole mass given by $x^i = X_h^i/(X_h^a + X_h^c)$, $X_h^i = e^2/h \int ds_{h(e)}(v_{F,h(e)}^i)^2/\psi_{F,h(e)}^i$, where $\psi_{F,h(e)}^i$ is $i$ (= $a, c$) component of the Fermi velocity of holes (electrons) $\psi_{F,h(e)}$ [10]. The hole mass determined by the dHvA measurements is nearly isotropic $m_h^a \approx m_h^c \approx 13 m_0$ [11], where $m_h^a$ ($m_h^c$) is the mass of the hole along the $a$ ($c$) axis and $m_0$ is the free electron mass. The heavy electron mass is anisotropic and is estimated to be $m_e^c \approx 85 m_0$ and $m_e^a \approx 305 m_0$ from the large Sommerfeld coefficient in the heat capacity ($\gamma \sim 80$ mJ/K$^2$mol) [2] and the anisotropy of $H_{c2}$ [12]. Then we obtain $x^a \approx 0.87$ and $x^c \approx 0.95$. Using these parameters we calculate the superfluid density by assuming the nodal topologies $\Delta_h(k, T) = \Delta_h(T) \times 2 \sin \theta \cos \theta$ with point and horizontal line nodes for hole bands and $\Delta_e(k, T) = \Delta_e(T) \times \sin \theta$ with $c$-axis point nodes for electron bands, which have been suggested by the thermal conductivity measurements [3].

The lower critical fields $H_{c1}^{a,c}(T)$ are related to the superfluid density as $H_{c1}^{a,c} \propto n_s^a$ and $H_{c1}^{a,c} \propto \sqrt{n_s^a n_s^c}$. First we try to fit $H_{c1}^{a,c}(T)$, and the best fit is obtained by $\Delta_h(0) = 1.6 k_B T_c$ and $\Delta_e(0) = 4.0 k_B T_c$. As shown by the thick line in Fig. 2(a), the fitting result well reproduces...
the overall temperature dependence of \( H_{c1}^0(T) \). We note that this \( \Delta_h(0) \) value is close to \( \Delta_e(0) \sim h v_{F,h} / \pi \xi_h \sim 1.5 k_B T_c \), which is obtained from \( v_{F,h} \sim 2 \times 10^4 \text{ m/s} \) [11] and \( \xi_h \sim 25 \text{ nm} \). Here \( \xi_h = (\Phi_0 / 2 \pi \mu_0 H_{c2}^h)^{0.5} \) is the coherence length of the hole band estimated from the "virtual upper critical field" of the hole band \( \mu_0 H_{c2}^h \sim 0.5 \text{ T} \) [3]. In Fig. 2(b), \( H_{c1}^0(T) \) calculated by using the same \( \Delta_h(0) \) and \( \Delta_e(0) \) values is plotted by the thick line. In sharp contrast to \( H_{c1}^0(T) \), the calculation strongly deviates from the data. We tried to fit the data by assuming various \( \Delta_h(0) \) and \( \Delta_e(0) \) values, but could not reproduce the data, particularly the anomaly at \( T_Q \).

At the present stage of the study, the origin of the anomaly at \( T_Q \) is not clear. A possible explanation for this may be given by a peculiar vortex dynamics associated with chiral domains due to the multi-component superconducting order parameter with broken time reversal symmetry (TRS). As suggested in Ref. 13, magnetic fields penetrate inside through the domain wall even in the Meissner state. In fact, the superconducting order parameter with broken TRS having a form \( k_z (k_x \pm i k_y) \) has been proposed in URu$_2$Si$_2$ at low temperatures [3]. The order parameters in the forms \( k_x k_z \) and \( k_z k_y \) may have slightly different transition temperatures due to the hidden order, and then the time reversal symmetry will be broken below the lower temperature. This order parameter with broken TRS can produce domain walls along the \( c \) axis, which may give rise to a peculiar reduction of penetration field only for \( H \parallel c \).

![Image](https://via.placeholder.com/150)

4. Summary
In summary, we have determined accurately the lower critical fields of ultraclean URu$_2$Si$_2$. We find several distinct features, which has never been reported in crystals with low RRR values. Particularly, the remarkable kink structure in the temperature dependence of \( H_{c1} \) observed only for \( H \parallel c \) cannot be simply accounted for by the multiband nature. The observed unprecedented behavior demonstrates a highly unusual superconducting state of URu$_2$Si$_2$, which requires further theoretical and experimental investigations.

Acknowledgments
We thank D.F. Agterberg, E.H. Brandt, H. Ikeda, K. Machida and M. Sigrist for helpful discussions and V. Mosser for providing Hall sensors. This work was supported by Grant-in-Aid for the Global COE program “The Next Generation of Physics, Spun from Universality and Emergence”, Grant-in-Aid for Scientific Research on Innovative Areas “Heavy Electrons” (No. 20102006) from MEXT of Japan, and KAKENHI from JSPS.

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