Magnetism and superconductivity in strongly correlated CeRhIn$_5$

**Tuson Park$^{1,2}$ and J D Thompson$^{1,3}$**

$^1$ Los Alamos National Laboratory, Los Alamos, NM 87544, USA
$^2$ Department of Physics, Sungkyunkwan University, Suwon 440-746, South Korea
E-mail: tp8701@skku.edu and jdt@lanl.gov

*New Journal of Physics* **11** (2009) 055062 (16pp)
Received 22 December 2008
Published 29 May 2009
Online at http://www.njp.org/
doi:10.1088/1367-2630/11/5/055062

**Abstract.** Specific heat studies of CeRhIn$_5$ as functions of pressure and magnetic field have been used to explore the relationship between magnetism and unconventional superconductivity, both of which involve the 4f electron of Ce. Results of these studies cannot be understood as a simple competition for Fermi-surface states and require a new conceptual framework.

**Contents**

1. **Introduction**................................................. 2
2. **Experimental**................................................. 3
3. **Results**.................................................. 4
   3.1. Phase diagram........................................ 4
   3.2. Coexistence of magnetism and superconductivity below $P_1$.......................... 5
   3.3. Field-induced magnetism and superconductivity................................. 9
4. **Discussion**............................................... 11
5. **Summary**................................................ 14
Acknowledgments............................................. 15
References................................................... 15

---

$^3$ Author to whom any correspondence should be addressed.
1. Introduction

When Ce is added as a dilute magnetic impurity into a non-magnetic host, antiferromagnetic (AFM) exchange $J$ between band electrons of the host and the localized 4f electron of Ce$^{3+}$ produces a magnetic-singlet ground state in the zero-temperature limit. This many-body process, the Kondo effect, renormalizes the effective mass of itinerant charge carriers that is reflected in a large specific heat Sommerfeld coefficient per mole Ce and that scales as $\gamma \propto 1/T_K$ [1], where the Kondo temperature $T_K \approx E_F \exp[-1/2J N(E_F)]$ and $N(E_F)$ is the density of electronic states at the Fermi energy $E_F$ of the host [2]. The resulting Fermi volume of the interacting system is ‘large’, i.e. counts both band electrons of the host and the 4f electron of Ce. In contrast to the well-understood impurity limit, a periodic array of Kondo atoms is much more complex. In this dense limit, the Kondo effect as well as interactions, such as the indirect Ruderman–Kittel–Kasuya–Yosida (RKKY) interaction, between local moments are present. With both Kondo and RKKY interactions depending on $J$, their interplay can be tuned by chemical environment, magnetic field and pressure to give magnetically ordered, superconducting (SC) or paramagnetic ground states of the Kondo lattice [3]. If Kondo screening dominates, then the Fermi volume of the heavy quasiparticles also will be large [4]. The surface of this large Fermi volume is susceptible to instabilities, such as superconductivity and/or formation of a spin-density wave (SDW). CeCu$_2$Si$_2$, the first heavy fermion superconductor [5], is an example. Its large Fermi volume of itinerant heavy quasiparticles forms unconventional superconductivity, a SDW or both, depending on chemical stoichiometry [6]. In the sense that these broken symmetries are the consequence of a Fermi-surface instability, the same (itinerant) electrons are involved in both superconductivity and magnetism. On the other hand, if the interaction between local moments dominates, local-moment magnetic order develops in a ‘small’ Fermi volume that does not count the localized electrons, and the magnetic entropy in the ordered state will be close to that expected from the degeneracy of the localized electrons. CeCu$_2$Ge$_2$ is an example of the localized limit; localized 4f electrons order antiferromagnetically near 4 K in a well-defined crystal-field doublet manifold. CeCu$_2$Ge$_2$ becomes SC at very high pressures [7], where the Kondo effect dominates, and, consequently, magnetism and superconductivity arise from electrons with very different characters, one localized and the other itinerant.

The dense Kondo system UPd$_2$Al$_3$ has been proposed to exhibit both localized and itinerant characters of its three 5f electrons, with two of the 5f electrons being localized and ordering antiferromagnetically and one of the 5f electrons being itinerant and participating in coexisting unconventional superconductivity [8]. From a theoretical perspective, division of electronic orbitals into localized and itinerant components is possible depending on the competition between intra-atomic Coulomb interactions and anisotropic hybridization of 5f electrons [9]. Though this mechanism may account for the behavior of U’s 5f electrons in UPd$_2$Al$_3$, it remains to be shown that such competition could lead to simultaneously localized and itinerant characters of a single f electron. In addition to the relationship between magnetism and superconductivity, the issue of itinerant or localized character of electrons is important for interpreting the nature of quantum criticality that emerges as the Neel transition of a Kondo lattice is tuned to zero temperature. The Hertz–Millis–Moriya theory is constructed to account for quantum-critical behavior arising from a zero-temperature spin instability of a large Fermi volume [4], and, indeed, transport and thermodynamic properties of CeCu$_2$Si$_2$ that emerge as its spin-density transition is tuned to $T = 0$ can be interpreted in this framework [10]. On the
other hand, behaviors of the Kondo lattice systems YbRh$_2$Si$_2$ [11] and CeCu$_{6-x}$Au$_x$ [12] near their quantum-critical points have led to qualitatively different models that assume localized 4f electrons [4], and in contrast to CeCu$_2$Si$_2$, neither of these systems exhibits superconductivity near their quantum-phase transition [13]. The distinction, however, between localized and itinerant magnetic phases is not always straightforward, with some theories arguing that they are distinct phases and others for a continuous transition from localized to itinerant antiferromagnetism, i.e. from small to strong Kondo coupling, as a function of some tuning parameter that also accesses the quantum-critical regime.

CeRhIn$_5$ is an example of complexity posed by a Kondo lattice. At atmospheric pressure, localized 4f electrons in CeRhIn$_5$ order antiferromagnetically at 3.8 K. Studies of dilute Ce in LaRhIn$_5$ show that the Kondo-impurity temperature $T_K \approx 0.15$ K is more than one order of magnitude smaller than $T_N$ of CeRhIn$_5$ at atmospheric pressure [3], consistent with local moment order and the small Fermi surface deduced from de Haas–van-Alphen (dHvA) experiments [14]. Whereas, the small impurity $T_K$ would imply a huge Sommerfeld coefficient of $>35$ J mol$^{-1}$K$^{-2}$ and negligible Kondo compensation of the local moment, the experimental Sommerfeld coefficient for CeRhIn$_5$ above $T_N$ is about 0.4 J mole K$^{-2}$; magnetic entropy below $T_N$ is $\approx 0.3$ Rln2; and the order moment of 0.79 $\mu_B$ is reduced by about 10% from that of Ce in a crystal-field doublet without the Kondo effect [15, 16]. Together, these observations are inconsistent with naive expectations and suggest that electrons in the Kondo lattice are neither simply localized nor itinerant. This is reflected as well in the response of CeRhIn$_5$ to pressure. Modest pressure induces a phase of coexisting superconductivity and local-moment magnetic order that evolves at higher pressures to a SC state without magnetic order [17]–[19]. Applying a magnetic field, however, to this higher pressure SC state induces magnetic order that coexists with superconductivity to a critical pressure where the field-induced magnetic order is tuned to $T = 0$ [19, 20]. At this critical pressure, transport and thermodynamic properties exhibit strong deviations from those of a heavy Landau Fermi liquid [21], and dHvA frequencies of principal orbits increase sharply [14], as if signaling a transition from small-to-large Fermi volume, again emphasizing ambiguity in the nature of electrons responsible for superconductivity, magnetism and non-Fermi-liquid responses. As discussed below, specific heat studies of CeRhIn$_5$ as a function of temperature, pressure and magnetic field explicitly reveal this ambiguity and allow exploration of its consequences on the symmetry of the SC gap.

2. Experimental

Single crystals of CeRhIn$_5$ were grown from In flux as described elsewhere [15]. Powder x-ray diffraction confirmed the HoCoGa$_5$ tetragonal structure, which can be viewed as layers of CeIn$_3$ and RhIn$_2$ stacked sequentially along the c-axis. Electrical resistance measurements indicated the high quality of these crystals, with a resistivity ratio $\rho(300 \text{ K})/\rho(0 \text{ K}) > 400$ and a residual resistivity of $\approx 40 \text{ n}\Omega \text{ cm}$. Specific heat measurements were performed in a hybrid clamp-type pressure cell made of Be–Cu/NiCrAl in which there was a Teflon cup that contained a silicone-fluid pressure transmitting medium, a small piece of Sn, whose inductively measured SC transition served to determine pressure in the cell at low temperatures, and a sample of CeRhIn$_5$. The specific heat was determined by ac calorimetry. In this technique, an alternating current through a heater, attached to one side of the sample, induced an oscillating change in temperature $T_{ac}$, detected on the other side of the sample by a calibrated Au–Fe thermocouple and that is inversely proportional to the sample’s specific heat $C \propto 1/T_{ac}$. This technique is not

New Journal of Physics 11 (2009) 055062 (http://www.njp.org/)
absolute because the amplitude and phase of temperature oscillations are influenced by heat transported to the surrounding pressure medium. Approximately absolute values of the specific heat of CeRhIn$_5$ were obtained by scaling the measured specific heat to that determined earlier by an adiabatic technique [22]. The pressure cell was attached to $^3$He cryostat that fit into a 9 T SC solenoid.

3. Results

3.1. Phase diagram

Figure 1 displays representative specific heat measurements of CeRhIn$_5$ under pressure and zero magnetic field. Specific heat data at 1.15 GPa, which are denoted by down triangles, show a magnetic transition to an incommensurate (ICM) AFM phase with $Q = (0.5, 0.5, 0.297)$ at 3.62 K and a pressure-induced SC transition at a much lower temperature ($T_c = 0.36$ K). With further increasing pressure, the magnetic transition temperature $T_N$ decreases, while $T_c$ progressively increases. At 2.05 GPa (circles), there is a large specific heat discontinuity at

![Figure 1](http://www.njp.org/)

Figure 1. (a) Specific heat of CeRhIn$_5$ as a function of temperature at zero magnetic field. $T_c$, which is assigned as the midpoint of the jump in specific heat due to superconductivity, is marked by arrows. (b) Temperature–pressure phase diagram at zero field. $T_c$ and $T_N$ are determined by specific heat measurements (filled symbols) and by electrical resistivity measurements (crosses).
2.24 K due to a SC phase transition, but evidence for long-range magnetic order is completely absent. Even though $T_N$ sensitively depends on pressure, the shape and full-width at half-maximum (FWHM) of the magnetic transition is almost independent of pressure, indicating that the nature of the magnetic transition has not been changed under these hydrostatic pressure environments. This conclusion is consistent with neutron-scattering measurements under pressure [16].

Under applied pressure, CeRhIn$_5$ has been shown to possess two critical points (see figure 1(b)) [19]. The first occurs at 1.75 GPa ($= P_1$), where the magnetic and SC transition temperatures become equal in the absence of an applied magnetic field and the second at $P_2$ where a field-induced magnetic transition is tuned to $T = 0$ [19, 20]. The magnetically ordered phase is completely suppressed for $P > P_1$; whereas magnetic and SC phases coexist on a microscopic scale for $P < P_1$ [17, 18]. The abrupt suppression of AFM at $P_1$ has been interpreted as an indication of a weakly first-order phase transition between a state in which magnetic order and superconductivity coexist to a purely SC phase [23]. When extrapolated below 1.15 GPa, the bulk $T_c$ determined from $C_p$ is zero near 0.5 GPa at which $T_N$ starts to decrease, indicating an interplay between magnetic and SC phases. This conclusion is consistent with our specific heat measurements that do not show any hint of bulk superconductivity above 70 mK at ambient pressure, but is different from [24] that claimed bulk superconductivity near 110 mK.

In figure 1(b), we also plot $T_c$ and $T_N$ determined from electrical resistivity measurements. At ambient pressure, there is a slight decrease in resistivity below 50 mK possibly due to incomplete filamentary superconductivity (not shown). Unlike the bulk $T_c$ that increases gradually with pressure, the $T_c$ determined from zero resistance sharply increases above 0.5 GPa, resulting in a large football-shaped difference curve in the two $T_c$s. For pressures above $P_1$, the two $T_c$s become almost equal. A difference between the bulk and transport $T_c$ also has been reported in a sister compound CeIrIn$_5$ [25]. Polishing, etching and heavy-ion irradiation of the surface of CeIrIn$_5$ do not affect the two $T_c$s, ruling out the possibility of surface superconductivity. Chemical substitution, however, narrows the difference between the two $T_c$s, suggesting that a strain may exist locally to affect superconductivity of CeIrIn$_5$ and chemical disorder relieves the local strain to make the system more homogeneous [26]. In CeRhIn$_5$, the difference between bulk and resistive $T_c$s is reversible with decreasing pressure, which indicates an intrinsic response to the coexistence of magnetic and SC orders and not an effect of internal strain. A strain scenario would require large pressure inhomogeneity, at least of order 0.5 GPa even at ambient pressure. Such a large pressure gradient is at odds with independent studies of the hydrostaticity of our pressure medium in which there is a maximum gradient of 0.01 GPa even when the silicone fluid freezes into a glassy state [27]. In addition, the coincidence of bulk and resistive transitions for $P > P_1$ is also inconsistent with a strain scenario. An alternative interpretation is that the resistive transition arises from magneto-elastic coupling that initially induces filamentary superconductivity at AFM domain walls before the sample bulk superconducts at lower temperature.

3.2. Coexistence of magnetism and superconductivity below $P_1$

Though specific heat measurements establish two phase transitions, bulk superconductivity and magnetic order, at pressures below $P_1$, and nuclear magnetic resonance (NMR) measurements show that these orders coexist on the scale of a unit cell, neither is able to determine the nature
of the electronic state out of which these orders develop. Neutron diffraction at pressures to 1.6 GPa show that the size of the ordered moment (0.79 \(\mu_B\)) is only weakly pressure dependent and close to that of Ce\(^{3+}\) in a crystal field doublet, indicating that magnetism in CeRhIn\(_5\) arises from localized Ce 4f spins [16]. The localized nature of Ce’s 4f electron in the AFM state is consistent with dHvA experiments on Ce-doped LaRhIn\(_5\), where quantum-oscillation frequencies reflecting a small Fermi surface are independent of Ce-doping concentration up to stoichiometric CeRhIn\(_5\) [28]. These results argue for local-moment order mediated by the RKKY interaction. As a function of pressure, the dHvA frequencies of CeRhIn\(_5\) are unchanged for \(P < P_1\) [14], i.e. the f electron remains localized so that the coexistence of magnetic order and superconductivity should not be viewed as a simple competition for electronic states at the Fermi energy. On the other hand, inspection of specific heat data plotted in figure 1(a) shows that superconductivity develops in the AFM state in which \(C/T\) just above \(T_c\) is a substantial fraction of \(C/T\) just above \(T_N\), that is, Neel order has not dramatically suppressed the effective mass of quasiparticles that form Cooper pairs. Such enhanced effective mass must come from hybridization of f- and band electrons since there is no enhancement of the effective mass of quasiparticles in non-magnetic LaRhIn\(_5\) [28]. These observations lead to a dichotomy in which the 4f electron produces both local moment magnetic order and participates in creating heavy mass quasiparticles that form superconductivity. We have used specific heat measurements as a function of pressure and magnetic field to explore the relationship between magnetism and superconductivity in the coexistence region below \(P_1\).

Figure 2(a) displays specific heat data for CeRhIn\(_5\) at 1.4 GPa and in magnetic fields of 0, 1.1 and 5.5 T. In zero field, a magnetic transition occurs at 3.0 K and then a SC transition follows at 0.9 K. Superconductivity is rapidly suppressed by an applied magnetic field, while the magnetic order is robust. At 5.5 T, there are two closely spaced specific heat anomalies. The sharpness of the second peak at lower temperature is indicative of its first-order nature. Magnetic field dependence of \(C/T\) at 0.3 K is plotted in figure 2(b). There are two anomalies at 0.6 and 2.3 T that correspond to SC and field-induced magnetic transitions, respectively. From data such as these, we construct the field–temperature phase diagram plotted in figure 2(c) that summarizes the evolution of electronic phase of CeRhIn\(_5\) at 1.4 GPa. The \(B–T\) phase diagram at this pressure is essentially identical to that at 1 bar [29], except for the presence of superconductivity at low-temperatures and low-fields (shaded area). Neutron-diffraction measurements at 1 bar show that the zero-field transition is to an ICM structure (0.5, 0.5, 0.297) [30]; whereas, the low-temperature, field-induced transition is CM at (0.5, 0.5, 0.25) [31].

Even though the zero-field magnetic transition temperature is suppressed from 3.8 K at 1 bar to 3.0 K at 1.4 GPa, the critical field \(B^*\) that is required for the spin reorientation transition from ICM to CM is almost independent of pressure up to this pressure: \(B^* = 2.0\) T at 1 bar and \(2.2\) T at 1.4 GPa and 0 K.

Figure 3(a) shows specific heat at 1.62 GPa, which is close to the critical boundary \(P_1\) and where \(T_N\) is depressed to 2.5 K and \(T_c\) is increased to 1.4 K. Like results at 1.4 GPa, the specific heat anomaly at \(T_N\) splits into two peaks under magnetic field. However, the critical field \(B^*\) that is required for the spin reorientation transition from ICM to CM has jumped to 4.4 T, a factor of 2 larger than that at 1.4 GPa. At 1.71 GPa, even closer to \(P_1\), \(B^* = 6.7\) T [32]. The field \(B^*\) is almost constant (\(\approx 2\) T) up to 1.4 GPa, where the SC upper critical field \(B_{c2}\) is much less than \(B^*\). For \(P \geq 1.5\) GPa, where \(B_{c2}\) sharply increases above 2 T, \(B^*\) increases in step with the increase in \(B_{c2}\) and does not cross \(B_{c2}\), indicating that superconductivity coexists with the ICM phase but competes against the CM phase. Though \(B^*\) and \(T_N\) change quantitatively with pressure, the
Figure 2. Specific heat for $B \perp c$-axis at 1.4 GPa. (a) Specific heat divided by temperature ($C/T$) is plotted as a function of temperature for 0, 1.1 and 5.5 T. (b) Magnetic field dependence of $C/T$ at 0.3 K. Arrows indicate SC and magnetic transitions at 0.66 and 2.3 T, respectively. Data were taken with increasing field. (c) $B$–$T$ phase diagram. Tuning magnetic field and temperature reveal three distinct magnetic phases of ICM with $Q = (0.5, 0.5, 0.297)$, commensurate (CM) with $Q = (0.5, 0.5, 0.25)$ and another ICM (II) magnetic order [31]. Solid lines are guides to eyes.

relationship among magnetic phases remains unchanged from $P = 0$ to $P < P_1$, reflecting little change in the localized nature of the 4f electron over this pressure range; nevertheless, the SC transition temperature increases by nearly an order of magnitude. We note that CeRhIn$_5$ is not alone in exhibiting this apparent localized/itinerant dichotomy. Its isostructural relative CeCoIn$_5$ is a heavy-fermion superconductor with a ‘large’ Fermi surface [33]. Replacing approximately 1 at% In with Cd induces a phase of coexisting antiferromagnetism and unconventional superconductivity [34, 35], and in several respects the temperature–Cd doping phase diagram is a mirror image $T$–$P$ phase diagram of CeRhIn$_5$. Neutron-diffraction measurements on 1 at% Cd in CeCoIn$_5$ find magnetic scattering intensity developing below $T_N$ that is arrested with the onset of superconductivity [36], clearly demonstrating a coupling between the two orders. It would be interesting to see if this same coupling were apparent in CeRhIn$_5$ at pressures less than $P_1$ and if the Fermi surface of Cd-doped CeCoIn$_5$ were ‘small’.

New Journal of Physics 11 (2009) 055062 (http://www.njp.org/)
Figure 3. Specific heat for $B \perp c$-axis at 1.62 GPa. (a) Specific heat divided by temperature ($C/T$) is plotted as a function of temperature for 0, 1.1, 4.4 and 6.6 T. (b) Magnetic field dependence of $C/T$ at 0.3 K. Arrows indicate SC and magnetic transitions at 0.66 and 2.3 T, respectively. The solid line is a least-square fit to $C/T = \gamma + \beta B^n$ where the best result was obtained with $n = 0.75$. (c) $B$–$T$ phase diagram based on the specific heat measurements.

Magnetic field dependence of the density of states (DOS) can be used to explore the nature of the SC order parameter. In conventional superconductors, where the SC gap is finite on the whole Fermi surface, the DOS is proportional to magnetic field because the DOS comes from states inside the normal core of vortices and the density of magnetic vortices is proportional to $B$. In unconventional superconductors with nodes, where the SC gap becomes zero on parts of the Fermi surface, the DOS mainly comes from extended quasiparticle states along the nodal directions and is expected to show a $B^{1/2}$ dependence [37]. Figure 3(b) shows $C/T$ as a function of magnetic field at 1.62 GPa. There are two anomalies that are associated with SC and spin-reorientation transitions at 3.4 and 4.6 T, respectively. The solid line is a least-squares fit to a power law, $C/T = \gamma + \beta B^n$, in the SC state, where $n = 0.75$ produces the best fit. The obtained exponent at this pressure does not conform to either a d-wave or an s-wave scenario. It is instructive to note that the SC upper critical field at this pressure, as shown in figure 3(c), also deviates from the conventional Werthamer–Helfand–Hohenberg (WHH) model of orbital pair breaking [38], showing a linear temperature dependence down to the lowest experimental
Figure 4. (a) Specific heat measurements at 1.88 GPa and for $B \perp c$-axis. $C/T$ is plotted as a function of temperature for 0, 2.2, 4.4 and 7.7 T. Arrows indicate a $B$-induced magnetic transition temperature. (b) Magnetic field dependence of $C/T$ at 0.3 K. (c) Field–temperature phase diagram of CeRhIn$_5$ at 1.88 GPa.

temperature. The nonconformity of the field-dependent specific heat and the temperature-dependent $B_{c2}$ may stem from the presence of ICM magnetic ordering, which could allow a SC order parameter other than spin-singlet Cooper pairing.

3.3. Field-induced magnetism and superconductivity

When pressure is higher than 1.75 GPa, evidence for a magnetic transition temperature $T_N$ is abruptly lost (see figure 1(b)), suggesting a first-order phase transition at $P_1$ from a coexisting phase of superconductivity and magnetism to a solely SC phase. The lack of magnetism above $P_1$ might be understood if conduction electrons required for the indirect RKKY interaction were completely gapped by the development of superconductivity; however, the SC gap is nonzero on parts of the Fermi surface at these pressures, as evidenced by a $T^3$ dependence of the nuclear relaxation rate [18]. Figure 4(a) displays specific heat measurements at 1.88 GPa that represent the high-pressure behavior of CeRhIn$_5$ for $P_1 < P < P_2$ (figure 1(b)). At zero field, a jump in the specific heat occurs at 2.2 K due to the onset of bulk superconductivity, but
there is no detectable feature that would indicate a magnetic transition at a lower temperature, e.g. near 1 K where a simple extrapolation of the Neel boundary would suggest that one might appear. When subjected to an external magnetic field as low as 1.1 T (not shown), however, a second peak in $C/T$ appears in the SC state. This low-temperature anomaly is enhanced with increasing magnetic field: the field-induced transition temperature $T_N$ increases and the peak anomaly becomes more conspicuous with increasing field, as expected if the anomaly signaled the development of long range magnetic order. It could be speculated that the field-induced transition in the SC state is similar to the spin-reorientation transition from ICM to CM state for $P < 1.75$ GPa. However, the fact that the spin-reorientation transition $B^*$ is larger than 6 T near 1.7 GPa but a field $B_M$ as low as 1.1 T is sufficient to induce magnetism at 1.88 GPa indicates that the two critical fields $B^*$ and $B_M$ are of different natures.

Figure 4(b) shows $C/T$ as a function of magnetic field at 0.3 K and 1.88 GPa. The lack of feature at the field-induced transition $B_M$ (1 T) is due to an immeasurably small change in specific heat at the transition in these field-swept data. As seen in these data, the field dependence of $C/T$ is very different from that at 1.62 GPa, now showing a linear $B$ variation. Even though the linear dependence is consistent with an s-wave order parameter, it is premature to conclude that a dramatic change in the nature of superconductivity has occurred upon crossing the critical pressure $P_1 = 1.75$ GPa because, as noted above, a $T^3$ dependence of $1/T$ in zero field implies a d-wave order parameter for $P > P_1$. Instead, the linear $B$ dependence may be influenced by the presence of field-induced magnetic order in the Abrikosov state, but to our knowledge there is no theoretical prediction for what might be expected in this case.

The relation between superconductivity and field-induced magnetic order, deduced from data such as shown in figure 4(a), is plotted in figure 4(c) and is characteristic of this relationship for pressures greater than $P_1$ but less than $P_2$. As seen here, the phase boundary $B_M(T)$ extends into the normal state above $B_{c2}$. It is interesting that the upper critical field boundary becomes linear in field just below the temperature where $B_M(T)$ crosses $B_{c2}(T)$ and that $B_M(T)$ has qualitatively different temperature dependences in and out of the SC state, both suggesting a coupling of the two order parameters. The near-field-independence of $B_M(T)$ above $B_{c2}(T)$ is reminiscent of the response of $T_N$ to field for $P < P_1$. This similarity further suggests that the field-induced magnetic order above $P_1$ is of the local-moment type, probably ICM, that is found below $P_1$. Neutron-diffraction studies would be very useful to confirm or deny this speculation. We note that the relation between field-induced magnetic order and superconductivity found here is distinctly different from what is observed in CeCoIn$_5$ where field-induced spin-density order exists only inside its Abrikosov phase [39].

At pressures higher than 2.35 GPa ($= P_2$), the field-tuned quantum-critical point [19], evidence for field-induced magnetic order disappears. This is illustrated in figure 5(a) where we plot specific heat data as a function of temperature for CeRhIn$_5$ at 2.45 GPa, which is slightly above $P_2$. With increasing field, $T_c$ is initially suppressed at a rate of 45.5 mK/T$^{-1}$ and at the highest field, $C/T$ continues to increase to the lowest temperatures without the appearance of a field-induced anomaly. If the upper critical field were determined by orbital pair-breaking effects, then the zero-temperature upper critical field, estimated from $B_{c2}(0) = -0.73T_c(dB_{c2}/dT) = 35$ T, far exceeds the experimental value of 7.3 T (see figure 5(c)). In addition to orbital pair breaking, in a spin-singlet superconductor, the upper critical field can be limited by field-induced alignment of spins, namely Pauli paramagnetic pair breaking [40]. The fact that the observed $B_{c2}$ is smaller by a factor of 5 than the estimated orbital critical field indicates that Pauli pairing breaking dominates, which contrasts with results below $P_1$.
Figure 5. Specific heat measurements at 2.45 GPa and for $B \perp c$-axis. (a) $C/T$ as a function of temperature for several fields. (b) $C/T$ at 0.3 K plotted as a function of magnetic field. (c) Field–temperature phase diagram. The dashed line is a linear fit to $B_{c2}$ near $T_c$ and gives a slope of 22 T K$^{-1}$.

(for example, figure 3(c)) where the extrapolated $B_{c2}(0)$ is larger than that determined by orbital effects. As shown in figure 5(b), the specific heat anomaly at $B_{c2}(0.3 \text{ K})$ is nearly symmetric, typical of a first-order phase transition, and in contrast to the second-order like anomaly at $T_c(B)$ in lower fields (figure 5(a)). Such a first-order transition is expected for a Pauli-limited $B_{c2}$ [41]. In this limit, we might expect the emergence of an inhomogeneous SC phase predicted by Fulde and Ferrell [42] as well as by Larkin and Ovchinnikov [43]. Though evidence for such a phase has been found in CeCoIn$_5$ [44, 45], which has a similar ratio of Pauli to orbital critical fields, we have not yet resolved a feature in the high-field, low-temperature specific heat of CeRhIn$_5$ that might be associated with this inhomogeneous SC phase.

4. Discussion

In conventional magnetic superconductors where magnetism and superconductivity coexist, localized f electrons from a rare-earth element contribute to a local magnetic order but
decoupled itinerant electrons are responsible for conventional superconductivity. In strongly correlated systems, in contrast, the same electrons can be responsible for both magnetism and unconventional superconductivity: Cu 3d electrons in high-$T_c$ cuprates or 4f (5f) electrons from rare-earth (or actinide) elements in heavy-fermion superconductors. Recently discovered Fe-pnictide superconductors also may belong to the class of electronically correlated superconductors (for example [46]), where Fe 3d electrons are responsible for both superconductivity and magnetic order (for example [47]).

Understanding the coexisting phase of CeRhIn$_5$ is complex even though the low-pressure AFM state and high-pressure SC state can be interpreted as arising from dominantly localized and itinerant characters of Ce’s 4f electron, respectively. The abrupt increase in dHvA frequencies near 2.35 GPa of a dominant quasi-two dimensional sheet [14] suggests a localized to delocalized transition in the 4f character, perhaps signaling a selective Mott transition at this quantum critical point [48]. Because all Fermi-surface sheets are not accounted for in these studies, it is not possible to conclude that the 4f electron has become completely delocalized to form a large Fermi surface at high pressures; however, these higher frequencies do correspond closely to those of the isostructural, heavy-fermion superconductor CeCoIn$_5$ whose Fermi-surface topology and size agree well with band calculations that assume itinerancy of the 4f electrons [49].

In the absence of a direct probe of the 4f electron, inferences of its contributions to magnetism and superconductivity are provided by the evolution of magnetic entropy with pressure—see figure 6. At ambient pressure, there is a sharp kink at $T_N$ (marked by an arrow) and about 70% of the Rln2 spin entropy is recovered near 8 K, where neutron measurements show the onset of short range spin–spin correlations [50]. Even though the ground state of CeRhIn$_5$ is sensitive to pressure, the entropy below 8 K is almost independent of the applied pressure and ground state. Figure 6(b) shows how entropy at $T_c$ and $T_N$ evolves as a function of pressure. At 1.05 GPa, entropy of the Neel order dominates, and entropy associated with superconductivity is negligible. With increasing pressure, entropy due to magnetic order decreases and the ‘lost’ entropy is transferred to SC entropy. When pressure is higher than 1.75 GPa, entropy is completely in the SC channel. The transfer of spin entropy from magnetism to superconductivity provides strong evidence that the single Ce 4f electron does participate in the two disparate broken symmetries and, further, that superconductivity has a magnetic origin.

The SC order parameter in the high-pressure phase of CeRhIn$_5$ is most likely d-wave. NMR measurements show a $T^3$ dependence below $T_c$ in the spin–lattice relaxation rate [18]; there is no Hebel–Slichter peak below $T_c$; the upper critical field is Pauli limited; and, as shown in figure 7, field-rotation specific heat measurements under pressure find a fourfold modulation when magnetic field is rotated within the Ce–In plane. We note that these pieces of experimental evidence also characterize CeCoIn$_5$, where a d-wave order parameter is established [51]–[53].

In the coexisting phase, the nature of the SC order parameter is less clear. The presence of magnetism as well as proximity to a magnetic critical point has prompted various theoretical predictions, ranging from a generalized d-wave with eight nodal points to a gapless p-wave superconductivity [54, 55]. Experimentally, NQR measurements show that $1/T_1$ crosses over from a $T^3$ to $T$-linear dependence deep in the SC state at 1.6 GPa, possibly indicating a gapless p-wave order parameter. Specific heat measurements, however, observe a finite $T = 0$ DOS in the coexisting phase, which can also lead to a $T$-linear crossover. In addition, field-rotation specific heat measurements have revealed a fourfold oscillation, which is consistent with a
Figure 6. (a) Entropy as a function temperature for several pressures at zero magnetic field. Inset: magnification of the low-T SC transition. (b) Comparison of entropy values at $T_N$ (solid squares) and $T_c$ (solid circles) as a function of pressure. Hashed area marks a zero-field critical pressure $P_1 (=1.75 \text{ GPa})$.

d-wave order parameter in the coexisting phase [56]. The dependence on magnetic field of $C/T$ in the coexisting phase, however, cannot be explained by a d-wave alone, requiring an interplay between magnetism and superconductivity.

As suggested from the evolution of entropy, unconventional superconductivity appears to have a magnetic origin. A phenomenological model of magnetically mediated superconductivity derives an attractive pairing potential that increases with the generalized momentum- and frequency-dependent magnetic susceptibility [57]. Because the magnetic susceptibility diverges at a magnetic quantum-phase transition, the pairing potential will be strongest there, and, indeed, superconductivity in heavy-electron systems typically emerges in proximity to a quantum-phase transition. The nature, however, of the quantum criticality should influence the dominant pairing channel. For example, the susceptibility is singular at the $Q$ vector defining a SDW that becomes critical at $T = 0$, and this favors d-wave pairing [57]. On the other hand, ferromagnetic criticality favors the p-wave channel. The critical state that emanates from $P_2$ in CeRhIn$_5$ is neither that of an SDW nor of ferromagnetism but is associated with criticality of localized 4f electrons [21]. Such criticality is consistent with a selective Mott or Kondo-breakdown type of zero-temperature phase transition [48], [58]–[61]. Unlike an SDW quantum-phase...
Figure 7. In-plane, field-angle-dependent ac specific heat divided by temperature for CeRhIn$_5$ at 2.3 GPa. Measurements are made at a fixed temperature of 325 mK and fixed magnetic field of 0.9 T, which is well below the upper critical field. The solid line is a least-squares fit to a fourfold modulation model: $C(\phi) = C(0) + A(1 - A4 \cos(4\phi)) + B\phi$, where $C(0) = 200$, $A = 9.3$, $A4 = 0.0856$ and $B = -0.00243$. We tentatively attribute the linear-in angle, $B\phi$, contribution to slight misalignment of the crystal.

transition, fluctuations emerging from this type of unusual quantum-phase transition are not well elaborated but are not expected to be peaked around a particular $\mathbf{Q}$. In this case, it is not clear what pairing channel might be favored or, from a theoretical perspective, even if superconductivity should appear. Nevertheless, CeRhIn$_5$ appears to provide an example of where this is possible. The simultaneous localized/itinerant roles of the 4f electron in CeRhIn$_5$ seem to be necessary ingredients in this physics.

5. Summary

Specific heat studies of the heavy-fermion compound CeRhIn$_5$ reveal a complex interplay between localized and itinerant degrees of freedom of Ce’s 4f electron. The evolution of this interplay with applied pressure is emphasized in the response of CeRhIn$_5$ to applied magnetic field which shows that the 4f electron remains localized up to a critical pressure $P2$. In spite of its local character, the 4f electron also participates in creating a state of bulk unconventional superconductivity. There are many open problems posed by these discoveries, not the least of which is the need for a theoretical basis for interpreting experimental observations. Understanding the physics of CeRhIn$_5$ should open new perspectives on other complex systems in which electronic correlations entangle states to create emergent functionalities.

New Journal of Physics 11 (2009) 055062 (http://www.njp.org/)
Acknowledgments

We thank H O Lee, F Ronning and V A Sidorov for communicating unpublished results and acknowledge numerous helpful discussions with colleagues. TP acknowledges a grant from the Korea Science and Engineering Foundation (KOSEF) funded by the Korea government R01-2008-000-10570-0. Work at Los Alamos was performed under the auspices of the US Department of Energy/Office of Science and funded in part by the Los Alamos Laboratory Directed Research and Development program.

References

[1] Bader S D, Phillips N E, Maple M B and Luengo C A 1975 Solid State Commun. 16 1263
[2] For example, Hewson A C 1993 The Kondo Problem to Heavy Fermions (Cambridge: Cambridge University Press)
[3] Yang Y-f, Fisk Z, Lee H O, Thompson J D and Pines D 2008 Nature 454 611
[4] For example, Lohneysen H v, Rosch A, Vojta M and Wolfle P 2007 Rev. Mod. Phys. 79 1015
[5] Steglich F et al 1979 Phys. Rev. Lett. 43 1892
[6] Grewe N and Steglich F 1991 Handbook on the Physics and Chemistry of Rare Earths vol 14 (Amsterdam: Elsevier) p 343
[7] Jaccard D, Wilhelm H, Alami-Yadri K and Vargoz E 1999 Physica B 259–261 1
[8] Caspary R et al 1993 Phys. Rev. Lett. 71 2146
[9] Efremov D V, Hasselmann N, Runge E, Fulde P and Zwicknagl G 2004 Phys. Rev. B 69 115114
[10] Steglich F et al 1996 J. Phys.: Condens. Matter 8 9909
[11] Paschen S et al 2004 Nature 432 881
[12] Schroder A, Aeplli G, Bucher E, Ramazashvili R and Coleman P 1998 Phys. Rev. Lett. 80 5623
[13] Gegenwart P, Si Q and Steglich F 2008 Nat. Phys. 4 186
[14] Shishido H et al 2005 J. Phys. Soc. Japan 74 1103
[15] Hegger H et al 2000 Phys. Rev. Lett. 84 4986
[16] Llobet A et al 2004 Phys. Rev. B 69 024403
[17] Mito T et al 2003 Phys. Rev. Lett. 90 077004
[18] Kawasaki S et al 2003 Phys. Rev. Lett. 91 137001
[19] Park T et al 2006 Nature 440 65
[20] Knebel G, Aoki D, Braithwaite D, Salce B and Flouquet J 2006 Phys. Rev. B 74 020501
[21] Park T et al 2008 Nature 456 366
[22] Fisher R A et al 2002 Phys. Rev. B 65 4509
[23] Flouquet J 2005 arXiv:cond-mat/0501602
[24] Paglione J et al 2008 Phys. Rev. B 77 100505
[25] Petrovic C et al 2001 Europhys. Lett. 53 354
[26] Bianchi A et al 2001 Phys. Rev. B 64 220504
[27] Sidorov V A unpublished
[28] Harrison N et al 2004 Phys. Rev. Lett. 93 186405
[29] Cornelius A L et al 2001 Phys. Rev. B 64 144411
[30] Bao W et al 2000 Phys. Rev. B 62 R14621
[31] Raymond S, Ressouche E, Knebel G, Aoki D and Flouquet F 2007 J. Phys.: Condens. Matter 19 242204
[32] Park T, Graf M J, Boulakovski L, Sarrao J L and Thompson J D 2008 Proc. Natl Acad. Sci. USA 105 6825
[33] Shishido et al 2002 J. Phys. Soc. Japan 71 162
[34] Pham L D, Park Tuson, Maquilon S, Thompson J D and Fisk Z 2006 Phys. Rev. Lett. 97 056404
[35] Urbano R et al 2007 Phys. Rev. Lett. 99 146402
[36] Nicklas M et al 2007 Phys. Rev. B 76 052401

New Journal of Physics 11 (2009) 055062 (http://www.njp.org/)
[37] For example, Sigrist M and Ueda K 1991 Rev. Mod. Phys. 63 239
[38] Werthamer N R, Helfand E and Hohenberg P C 1966 Phys. Rev. 147 295
[39] Kenzelmann M et al 2008 Science 321 1652
[40] Clogston A M 1962 Phys. Rev. Lett. 9 266
[41] Maki K 1966 Phys. Rev. 148 362
[42] Fulde P and Ferrell R A 1964 Phys. Rev. 135 A550
[43] Larkin A I and Ovchinnikov Y N 1964 Zh. Eksp. Teor. Fiz. 47 1136
Larkin A I and Ovchinnikov Y N 1965 Sov. Phys.—JETP 20 762 (Engl. Transl.)
[44] Bianchi A, Movshovich R, Capan C, Pagliuso P G and Sarrao J L 2003 Phys. Rev. Lett. 91 187004
[45] Radovan H et al 2003 Nature 425 51
[46] Haule K, Shim J H and Kotliar G 2008 Phys. Rev. Lett. 100 226402
[47] Christianson A D et al 2008 Nature 456 930
[48] Pepin C 2007 Phys. Rev. Lett. 98 206401
[49] Haga Y et al 2001 Phys. Rev. B 63 060503
[50] Bao W et al 2002 Phys. Rev. B 65 100505
[51] Movshovich R et al 2001 Phys. Rev. Lett. 86 5152
[52] Izawa K et al 2001 Phys. Rev. Lett. 87 057002
[53] Park W K, Sarrao J L, Thompson J D and Greene L H 2008 Phys. Rev. Lett. 100 177001
[54] Bang Y, Graf M J, Balatsky A V and Thompson J D 2004 Phys. Rev. B 69 014505
[55] Fuseya Y, Kohno H and Miyake K 2003 J. Phys. Soc. Japan 72 2914
[56] Park T, Bauer E D and Thompson J D 2008 Phys. Rev. Lett. 101 177002
[57] Monthoux P, Lonzarich G G and Pines D 2007 Nature 450 1177
[58] Coleman P, Pepin C, Si Q and Ramazashvili R 2001 J. Phys.: Condens. Matter 13 R723
[59] Si Q, Rabello S, Ingersent K and Smith J L 2001 Nature 413 804
[60] Senthil T, Sachdev S and Vojta M 2003 Phys. Rev. Lett. 90 216403
[61] Paul I, Pepin C and Norman M R 2007 Phys. Rev. Lett. 98 026402

New Journal of Physics 11 (2009) 055062 (http://www.njp.org/)