Probing the Nodal Gap in the Pressure-Induced Heavy Fermion Superconductor CeRhIn$_5$

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We report field-orientation specific heat studies of the pressure-induced heavy-fermion superconductor CeRhIn$_5$. These experiments provide the momentum-dependent superconducting gap function for the first time in any pressure-induced superconductor. In the coexisting phase of superconductivity and antiferromagnetism, field rotation within the Ce-In plane reveals fourfold modulation in the density of states, which favors a d-wave order parameter and constrains a theory of the interplay between superconductivity and magnetism.

The emergence of superconductivity near an antiferromagnetic quantum critical point (QCP) suggests that magnetic fluctuations associated with the QCP may provide a glue to form electron pairs [1]. In unconventional superconductors, the superconducting (SC) order parameter, which is tied closely to the SC pairing mechanism, breaks the underlying crystalline symmetry and contains nodes at which the SC gap becomes zero on the Fermi surface. The presence of nodal quasiparticles qualitatively changes the density of states (DOS) at the Fermi level and leads to a power-law temperature dependence of thermodynamic properties [2]. This power-law behavior contrasts with the exponential temperature dependence in fully gapped conventional superconductors and, therefore, provides an indicator of unconventional superconductivity. Unambiguous determination of an unconventional SC order parameter, however, comes from phase-sensitive and momentum-dependent studies [3].

In heavy-fermion superconductors, where the relationship between superconductivity and quantum criticality is most conspicuous [1], details of the SC gap are rarely available because of disorder from chemical substitution or pressure environments that are needed to induce superconductivity. In this Letter, we report field-orientation specific heat (C_p) measurements on the pressure-induced superconductor CeRhIn$_5$. When a magnetic field is rotated in the Ce-In plane, fourfold modulation in C_p is observed deep in the SC state, with minima along [100] that indicate gap zeros at this direction in a d-wave order parameter. These results provide the momentum-dependent SC gap function for the first time, which is also the first in any pressure-induced superconductor, and open a new avenue to insights on the pairing mechanism in strongly correlated superconductors.

CeRhIn$_5$ is an antiferromagnet below $T_N = 3.8$ K and belongs to a family of heavy-fermion compounds CeMIn$_5$ ($M =$ Co, Ir, Rh) that become superconducting for $M =$ Co and Ir at 2.3 and 0.4 K, respectively [4–6]. Derived from the antiferromagnet CeIn$_3$, it crystallizes in the tetragonal HoCoGa$_5$ structure, where a Ce-In plane and a M-In block

are alternately stacked along the c axis. When subjected to pressure, CeRhIn$_5$ becomes superconducting near 0.5 GPa where T_N starts to decrease, suggesting a correlation between superconductivity and magnetism [see Fig. 1(a)] [4,7,8]. As shown in Fig. 1(a), there are two critical pressures at 1.75 and 2.3 GPa denoted $P1$ and $P2$. Below $P1$, superconductivity and magnetism coexists on a micro-
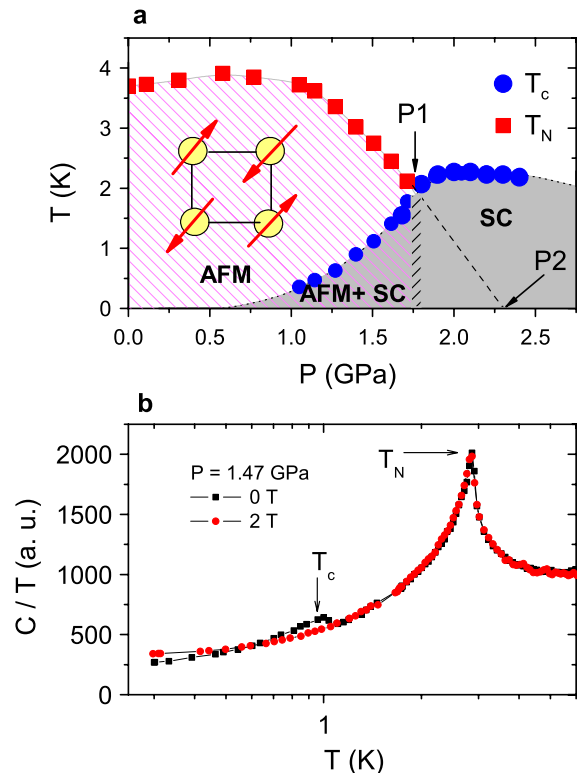

FIG. 1 (color online). (a) Phase diagram of CeRhIn$_5$ as a function of pressure [15]. Pressure induces a bulk superconducting (SC) state at 0.5 GPa that coexists with the antiferromagnetic (AFM) phase up to $P1$, where T_c becomes equal to T_N. The critical pressure marked by $P2$ is the field-tuned quantum critical point hidden by the SC dome. (b) Heat capacity of CeRhIn$_5$ at 1.47 GPa (see text for details).

scopic scale, while a purely SC phase only appears above
$P1$ [9]. $P2$ is a field-tuned quantum critical point hidden by
the SC dome [7]. At pressures above $P1$, the zero-field
nuclear spin-relaxation rate $1/T_1$ and specific heat show a
power-law temperature dependence below T_c, which is
similar to what is found in the ambient-pressure heavy-
fermion superconductor CeCoIn$_5$ with a d-wave SC order
parameter [9,10]. In the coexisting phase below $P1$, the
temperature dependence of $1/T_1$ crosses over from T^3 to
T-linear with decreasing temperature in the SC state, sug-
gesting a change from d-wave singlet pairing to an exotic
SC order parameter [11]. Singular quantum fluctuations in
the vicinity of an AFM QCP also have been proposed to
favor unconventional order parameters ranging from a
p-wave singlet to extended d-wave with additional nodal
points [12,13].

  Whereas power laws in $1/T_1$ and C_p are consistent with
a nodal gap in the coexistence phase, these measurements
are insensitive to the location of nodes. To resolve this
important question, we have developed a field-rotation
specific heat technique under pressure. Single crystals
used in this study were obtained from a batch where a
residual resistivity ratio (RRR) was measured routinely to
be of the order of 500, indicating exceptional sample
quality. A hybrid clamptype pressure cell made of
Be-Cu/NiCrAl and silicone-fluid transmitting medium
were used to produce hydrostatic pressure environments
up to 2.5 GPa. The superconducting transition temperature
of Sn was measured inductively to determine the pressure
at low temperatures. Magnetic field rotation was performed
by using a triple-axis vector magnet with a large bore of
$2.5''$ (American Magnetic Inc.) that accommodated a ^{3}He
cryostat with the pressure cell. Specific heat under pressure
was measured by an ac calorimetric technique where ac
heating incurs oscillation in the sample temperature (T_{ac})
that is inversely proportional to the sample's heat capacity,
i.e., $T_{ac} \propto 1/C$ [14,15].

  Figure 1(b) shows the temperature dependence of the
heat capacity of CeRhIn$_5$ in its coexisting phase at
1.47 GPa. Long-range magnetic order with wave vector
$\mathbf{Q} = (0.5, 0.5, 0.297)$ appears at 2.86 K (T_N) and is robust
against magnetic field, whereas the SC transition occurs at
1.04 K (T_c) and is completely suppressed at 2 T ($H \parallel a$
axis). The magnetic transition at this pressure is as sharp as
it is at ambient pressure, indicating a negligibly small
pressure gradient in the pressure cell.

  The low-T heat capacity of CeRhIn$_5$ is shown in
Fig. 2(a) for representative fields along [100] (solid sym-
bols) and along [001] (open symbols). These data allow a
determination of anisotropy of the upper critical field H_{c2}.
Within the ab plane, the upper critical field H_{c2}^{ab} is essen-
tially isotropic [see Fig. 2(b)], but H_{c2}^c along the c axis is
smaller by a factor of 2, which reflects the tetragonal
crystalline structure as found in CeCoIn$_5$ [16]. For mag-
netic field along any crystalline axis, H_{c2} reveals a linear-T
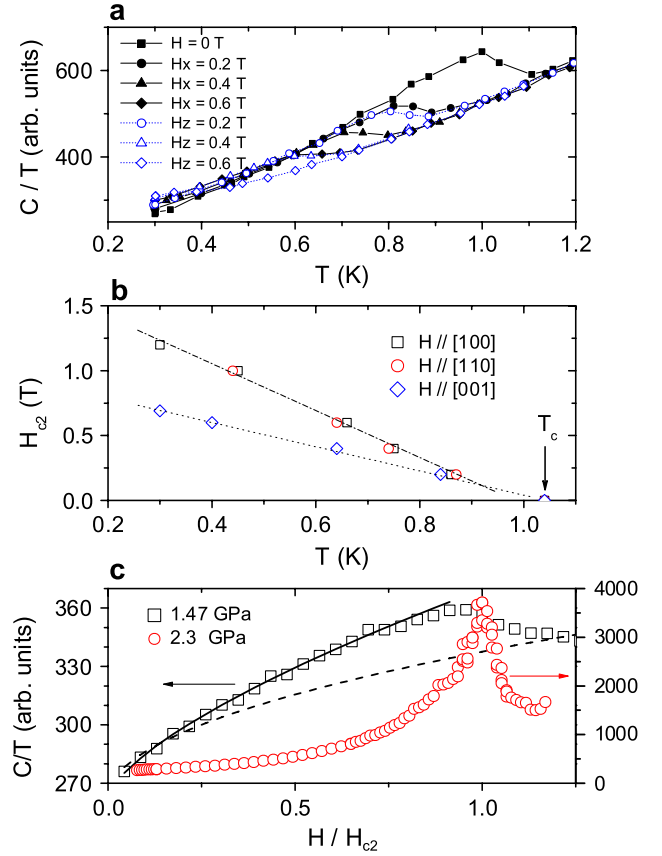dependence down to the lowest experimental temperature

FIG. 2 (color online). (a) Low-temperature heat capacity (C_p)
of CeRhIn$_5$ at 1.47 GPa and in low applied fields indicated in the
legend. Solid and open symbols represent C_P/T for field along
[100] and [001], respectively. (b) Temperature dependence of the
upper critical field H_{c2}. Squares, circles, and diamond symbols
correspond to fields along [100], [110], and [001], respectively.
The dotted (or dash-dotted) line is a guide for a linear-H
dependence. (c) Magnetic field dependence of C/T at 300 mK
for $H \parallel$ [100] at 1.47 GPa (squares) and 2.3 GPa (circles). Here,
H_{c2} is 1.2 and 7.7 T for 1.4 and 2.3 GPa, respectively. The solid
line describes least-squares fitting by $\Delta C/T = C/T -
C(0T)/T = AH^n$ with $n = 0.68$, while the dashed line is for
$n = 0.5$.

(0.3 K), which is different from CeCoIn$_5$ where Pauli
paramagnetic effects dominate conventional orbital fields
($H_{c2}^{orb} = -0.73 T_c dH_{c2}/dT$) and H_{c2} is smaller than H_{c2}^{orb}
by a factor of 5 [17]. Considering the exceptional sample
quality of CeRhIn$_5$ ($\rho_0 = 30$ n$\Omega \cdot$ cm), the lack of Pauli
limiting may arise from the presence of AFM that allows a
SC order parameter other than a spin singlet [18].

  Field dependence of the specific heat of CeRhIn$_5$ does
not conform to expectations for conventional type-II super-
conductors. Figure 2(c) shows representative field-swept
data at 300 mK for H along [100] at 1.47 GPa (squares) and
at 2.3 GPa (circles). In fully gapped conventional super-
conductors, C/T, which is proportional to the density of
states (DOS), linearly depends on magnetic field, while it
increases as $\sqrt{H}$ in unconventional superconductors with
nodes [19]. A power-law form of $C/T = C(0T)/T + AH^n$

was used to fit the field-dependent specific heat of CeRhIn$_5$ at 1.47 GPa, and the solid line in Fig. 2(c) best describes the data with $n = 0.68 \pm 0.04$ for field along [100]. This anomalous behavior may reflect the interplay between superconductivity and coexisting antiferromagnetism [15,20]. The field dependence of C/T at 2.3 GPa is strikingly different from that in the coexisting phase, showing a first-order like sharp feature at H_{c2}. The upper critical field of CeCoIn$_5$ also is first order below $T_c/2$, suggesting that CeCoIn$_5$ is similar to the high-pressure phase of CeRhIn$_5$ [7,21,22].

Figure 3 shows the field-angle specific heat of CeRhIn$_5$ at 1.47 GPa for field rotations ϕ (θ) within (perpendicular to) the Ce-In plane. In the paramagnetic state at 3 K, C/T is featureless as a function of angle (not shown). In the AFM state but above T_c [squares in Fig. 3(a)], C/T shows a broad feature around [010] that may reflect the presence of AFM domains below $T_N(= 2.86$ K). Upon decreasing
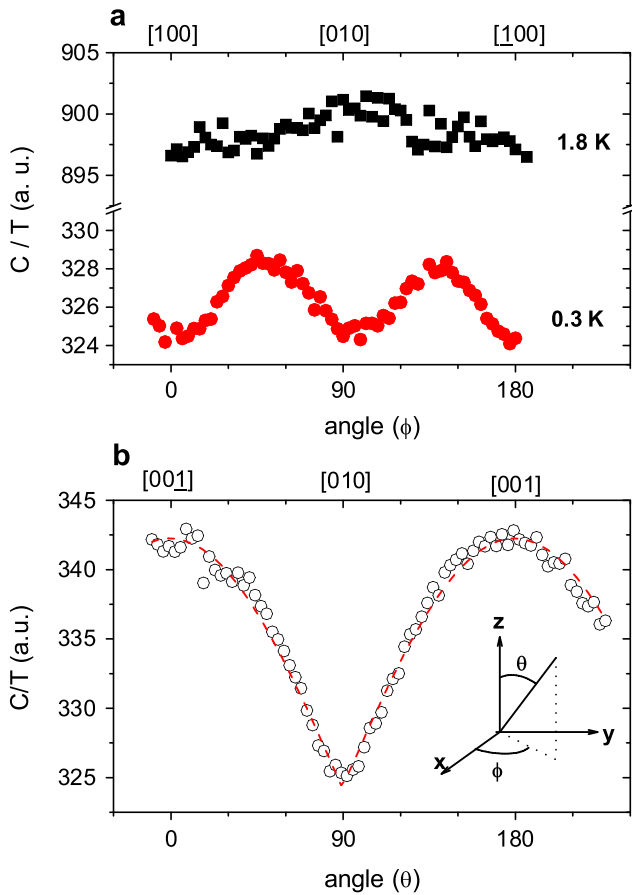

FIG. 3 (color online). Field-rotation specific heat of CeRhIn$_5$ at 1.47 GPa. (a) Azimuthal rotation (ϕ) of the magnetic field (0.5 T) within the Ce-In plane both in the AFM (1.8 K) and the coexisting phase of AFM and SC (0.3 K). (b) Polar rotation (θ) of the magnetic field (0.5 T) perpendicular to the Ce-In plane in the coexisting phase at 0.3 K. The solid line is a least-squares fit to a twofold modulation model: $C = C_0 + A(1 + A_2|\cos\phi|)$ with $C_0 = 265$, $A = 59.3$, and $A_2 = 0.30$, where the oscillation arises from H_{c2} anisotropy.

temperature below $T_c(= 1.04$ K), a new modulation in $C(\phi)/T$ appears in the mixed SC state [circles in Fig. 3(a)]. The fourfold modulation is distinctly different from the spectrum observed in the magnetic state, indicating that the new structure is not related to magnetism, but arises from the SC gap symmetry. A polar sweep at the same pressure, temperature, and field [Fig. 3(b)] shows only a twofold modulation that reflects the factor of 2 anisotropy in H_{c2} between the ab plane and c axis.

Field-rotation specific heat, which is proportional to the angle-dependent DOS at low temperature, has been used to probe the momentum-dependent SC gap function in anisotropic superconductors [22–27]. In the Abrikosov state, supercurrent circulates around a vortex core, and extended quasiparticles experience a Doppler energy shift, $\delta E \approx \mathbf{v_s} \cdot \mathbf{k_F}$, where $\mathbf{k_F}$ is the Fermi momentum of the quasiparticles and $\mathbf{v_s}$ is the supercurrent velocity. Derived from the Doppler effects that depend on the relative magnetic field orientation against nodal points on the Fermi surface, the DOS precisely reflects the anisotropic gap and modulates with field-rotation angle such that the DOS is minimal for field along nodal points but is maximal for field along antinodes [23]. Contributions to the field-rotation specific heat can be written as $C_{\text{total}}(\phi) = C_0 + C_2(\phi) + C_4(\phi)$, where the constant C_0 is the zero-field specific heat arising from thermally excited quasiparticles and phonons, the twofold $C_2(\phi)$ comes from sample misalignment, and $C_4(\phi)$ describes modulation from the SC order parameter. The field-angle ϕ was determined against the crystalline a axis or [100]. The lack of a twofold oscillation indicates that the ab plane is well matched with the field-rotation plane. Figure 4(a) is a plot of the field-induced heat capacity divided by temperature $\Delta C/T$ at 0.3 K and 1.47 GPa as a function of magnetic field angle, where $\Delta C = C_{\text{total}} - C_0 = C_4(\phi)$. Minima in ΔC can be approximated by a parabolic form $A(1 - A_4\cos4\phi)$ (dashed line) that is expected for the amplitude of a model gap structure of $\Delta(\theta, \phi) = \Delta_0\sqrt{1 + \cos4\phi}$ on a three-dimensional Fermi surface. Here, θ is a polar angle determined against the c axis.

Figure 4(b) shows the fourfold oscillation amplitude A_4 at 0.3 K and 1.47 GPa as a function of magnetic field, which is obtained by a least-squares fit to the sinusoidal form [dashed line in Fig. 4(a)]. Compared to negligible H_{c2} anisotropy within the ab plane [Fig. 2(b)], the large oscillation amplitude ($\approx 3.3\%$) in $\Delta C(\phi)$ of CeRhIn$_5$ indicates that the fourfold modulation is not due to Fermi surface anisotropy, but due to symmetry of the SC gap function [25,26]. Like CeCoIn$_5$, minima in ΔC of CeRhIn$_5$ are located along $\langle 100 \rangle$, indicating d_{xy} symmetry with line node along that direction [22,26]. Field-rotation measurements at 2.3 GPa and 0.5 T, where only superconductivity is observed, reveal fourfold in-plane modulation (not shown) similar to that at 1.47 GPa. In a recent theoretical treatment, however, Vorontsov and Vekhter argued that a reversal in the DOS anisotropy occurs at a critical field due
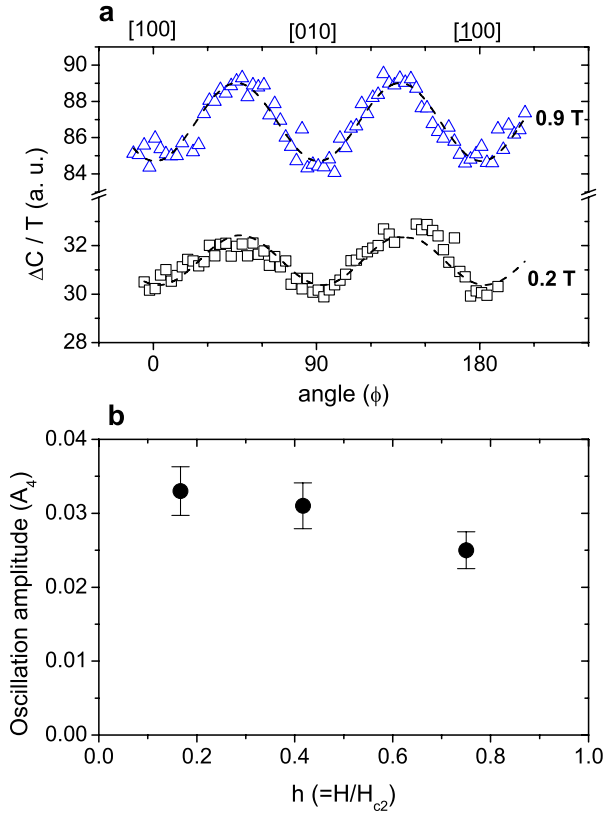
FIG. 4 (color online). (a) Field-rotation specific heat of CeRhIn$_5$ at 0.3 K for 0.2 (squares) and 0.9 T (triangles). Dashed lines are least-squares fits by a sinusoidal form of the angular dependence (see text). (b) Oscillation amplitude A_4 as a function of the normalized field $h(= H/H_{c2})$ with $H_{c2} = 1.2$ T at 0.3 K.

to additional pair breaking from vortex scattering; consequently, minima along $\langle 100 \rangle$ in C_p can be interpreted instead as evidence for a $d_{x^2-y^2}$ gap [28]. A key future experiment to distinguish the two d-wave order parameters is to identify the critical field at which anisotropy reversal occurs in the field-rotation specific heat of CeRhIn$_5$.

These results constrain possible candidates of the SC gap in the coexisting phase of antiferromagnetism and superconductivity of CeRhIn$_5$. Fourfold modulation of the in-plane specific heat excludes an extended d-wave with additional nodes at points where the antiferromagnetic Brillouin zone crosses the Fermi surface, which was suggested to account for the T-linear $1/T_1$ at lowest temperatures [13]. A gapless p-wave spin-singlet state that was predicted [12] to be favored over a d-wave symmetry near a QCP is also inconsistent with our observations. The twofold modulation in specific heat under polar field sweeps, as shown in Fig. 3(b), rules out the possibility of a hybrid gap with nodes at the poles.

To summarize, fourfold and twofold modulations in representative in-plane and out-of-plane field-rotation specific heat measurements on CeRhIn$_5$ favor a d-wave SC order parameter over other suggested candidates in the vicinity of the quantum critical point, thus constraining a theory of the interplay between superconductivity and antiferromagnetism. This work will assure further studies of other pressure-induced superconductors where the nature of the SC order parameter has yet to be identified.

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