Field-induced transition within the superconducting state of $CeRh_2As_2$

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In this report I want to highlight the discovery of two-phase unconventional superconductivity in $CeRh_2As_2$. Using thermodynamic probes, we establish that the superconducting critical field of its high-field phase is as high as 14 T, remarkable in a material whose transition temperature is 0.26 K. Furthermore, a c-axis field drives a transition between two different superconducting phases. In spite of the fact that $CeRh_2As_2$ is globally centrosymmetric, we show that local inversion-symmetry breaking at the Ce sites enables Rashba spin-orbit coupling to play a key role in the underlying physics. More detailed analysis suggests the transition from the low- to high-field states is associated with one between states of even and odd parity.

Unconventional superconductivity is usually associated with exotic, purely electronic pairing. Many of the electronic pairing mechanisms are expected to result in several superconducting states that are energetically close, and small changes of temperature, magnetic field and pressure might induce changes of the superconducting state. However, phase diagrams with multiple superconducting phases are rarely observed and most unconventional superconductors have simple phase diagrams, with a single superconducting state. Until now, the only stoichiometric superconductor that has been well established to have such a rich temperature - magnetic field superconducting phase diagram at ambient pressure is UPt_3 [1].

In our recent paper just accepted in Science [2], we report the discovery of a second such example, the heavy fermion material $CeRh_2As_2$ (https://www1.cpfs.mpg.de:2443/PQM_04). Our experimental data provided in the figures below show that it has extremely high superconducting critical fields of up to 14 T in spite of a superconducting transition temperature T_c of only 0.26 K. Furthermore, when the magnetic field is applied along the crystallographic c-axis, the superconducting state contains a well-defined internal phase transition at

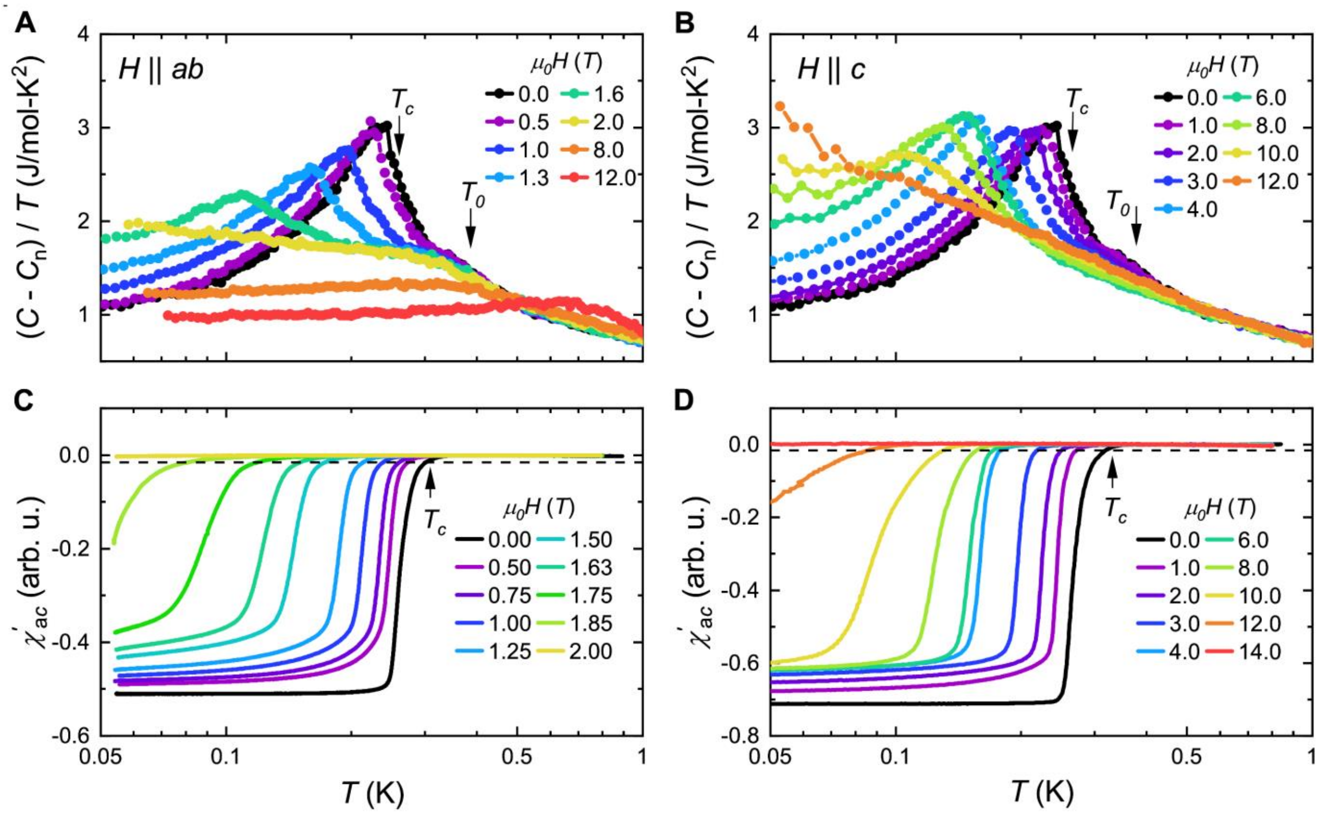

Fig. 1: Low-temperature phase transitions in $CeRh_2As_2$ and their magnetic field dependence. A and B show the specific heat for magnetic fields of different orientations. C and D show the magnetic ac-susceptibility. T_0 indicates a phase transition to an unknown ordered state whereas T_c indicates the onset of an unconventional superconducting state. [2]

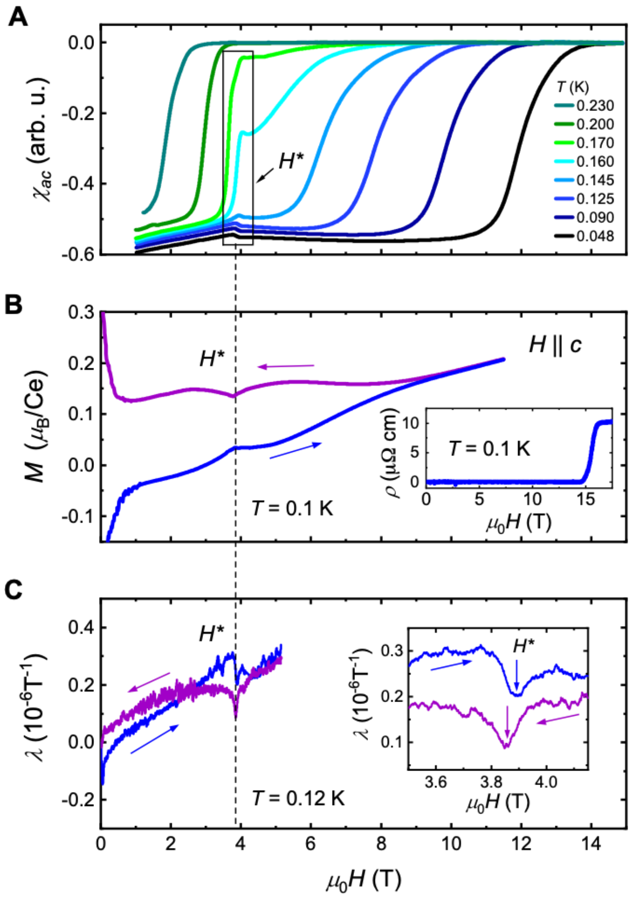

Fig. 2: Magnetic field dependence of the ac-susceptibility (A), the magnetization (B) and the magnetostriction (C) for H || c. The inset in (B) shows the resistivity and the inset in (C) zooms in on the data in (C) around the kink.

approximately 4 T that we identify using several thermodynamic probes. These remarkable features of superconductivity in $CeRh_2As_2$ are likely a manifestation of local inversion symmetry breaking and consequent Rashba spin-orbit coupling in an overall inversion-symmetric crystal structure.

$CeRh_2As_2$ is a heavy fermion system showing typical Kondo lattice behavior [2]. Below 1 K, two anomalies appear in the specific heat as shown in figure 1A and B for H = 0. A small hump is visible at $T_0 \approx 0.4$ K, marked by an arrow. It hints at a phase transition to an ordered state whose origin is yet to be determined, but the absence of an anomaly in the magnetic susceptibility at T_0 suggests that it might have Ce-4f multipolar or nematic character. In this report, we concentrate on the superconducting state experimentally evidenced by the large jump in the specific heat below 0.3 K. An equal entropy analysis reveals $T_c = 0.26$ K. The diamagnetic drop of the ac-susceptibility confirms entry to the superconducting state (figure 1C and D) at a similar T_c for the transition midpoint but a slightly higher onset temperature. In magnetic field, T_c shifts down and the superconducting transition is completely suppressed down to 0.05 K at 14 T for *H* || c and 2 T for *H* || ab. These are remarkably large critical fields for a superconductor with at T_c of only 0.26 K, especially for *H* || c. Temperature sweeps for *H* || c reveal a kink in the T_c (H) curve at the field of about 4 T, below which T_c is suppressed more quickly.

A pronounced kink in T_c (*H*) is suggestive of the existence of two superconducting phases. Indeed, this is confirmed by field sweeps of the ac susceptibility and two separate thermodynamic probes, magnetization and magnetostriction (figure 2). Remarkably, all three provide striking evidence of a phase transition. Below *T* = 0.2 K, pronounced kinks in all three observables are seen at a characteristic field, $H^* = 3.8$ T, that is almost temperature independent. As shown in figure 2A and the inset of figure 2B, diamagnetic shielding and zero resistivity persist in the vicinity of the phase transition, further proving that it occurs within the superconducting state. In contrast, field-dependent data for *H* ‖ ab show no sign of such a phase transition.

Using the values of T_c, H_{c2} and H^* from our measurements, we show the superconducting phase diagrams of $CeRh_2As_2$ for out-of-plane and in-plane fields in figure 3. From these phase diagrams the superconducting critical field can be extrapolated to $H_{c2}(0) \approx 14$ T for *H* || c and 1.9 T for *H* || ab. For *H* || c two superconducting states appear, labelled as SC1 and SC2, separated by a line that intersects the strong kink in the H_{c2}(T) curve in a multicritical point.

Looking more closely at the critical fields, we estimate the orbital limit from the slope of H_{c2} (T) near T_c via the Werthamer-Helfand-Hohenberg formula. This yields $H_{orb} \approx 17$ T for *H* || c and $H_{orb} \approx 8$ T for *H* || a, respectively. These estimates suggest that the upper critical field of SC2 along the c-axis is not Pauli-paramagnetically suppressed but the superconducting state SC1 is strongly Pauli limited with Pauli critical fields that are enhanced compared to the Clogston-Chandrasekhar limit of approximately 0.5 T for both field-directions.

Such a large anisotropy in the critical fields and large c-axis critical field is reminiscent of the non-centrosymmetric heavy fermions superconductors $CeCoGe_3$ $CeRhSi_3$ and $CeIrSi_3$ whose low crystal symmetry allows Rashba spin-orbit coupling [3]. Due

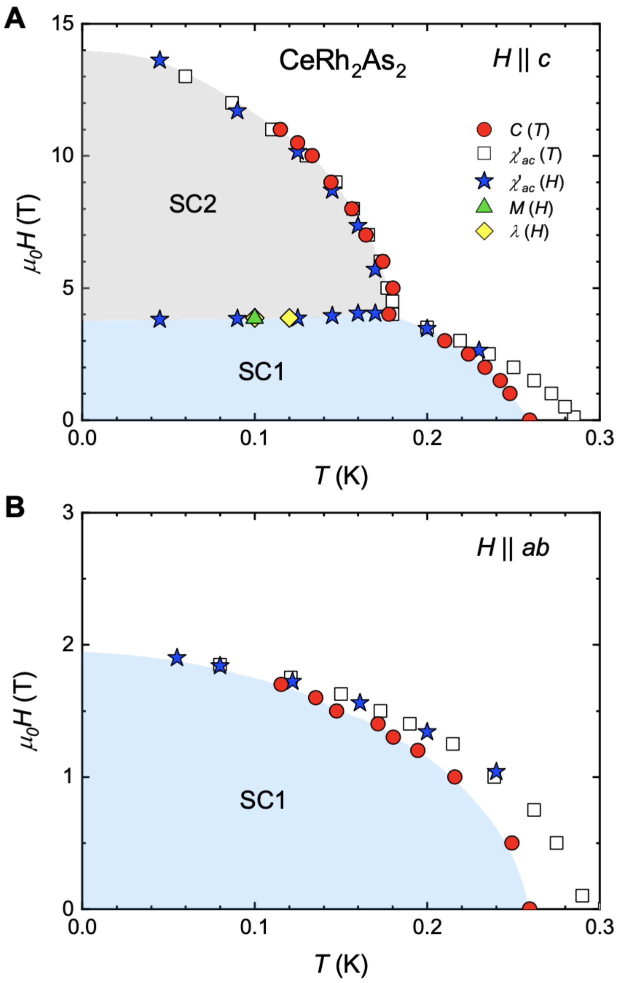

*Fig. 3: Superconducting phase diagram of $CeRh_2As_2$ for magnetic field applied along the c-axis (**A**) and in the plane (**B**) [2].*

to the broken inversion symmetry, even-parity (spin-singlet) and odd-parity (spin-triplet) superconducting states are not distinct and are in general mixed. These mixed states generally show no Pauli paramagnetic suppression for fields along the c-axis whereas they do for in-plane fields. However, these materials exhibit two important differences with respect to $CeRh_2As_2$: the first is that they do not have multiple superconducting phases; the second is that inversion symmetry is preserved in $CeRh_2As_2$. While $CeRh_2As_2$ is globally centrosymmetric (crystal structure in figure 4), Ce-square lattices, linked by the inversion symmetry, are themselves located in locally noncentrosymmetric environments, thus experiencing a Rashba interaction. A key feature of the centrosymmetric structure is that now even-parity (spin-singlet) and odd-parity (spin-triplet) Cooper pairs are not mixed, so a phase transition between even- and odd- parity condensates can occur. The even parity state is expected to be Pauli limited and therefore

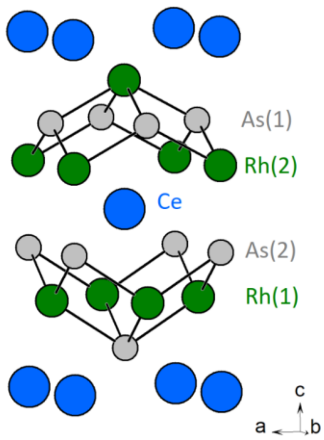

Fig. 4: Crystal structure of $CeRh_2As_2$.

more easily suppressed by magnetic field. Consequently, for higher fields, the odd parity state emerges, which itself is not Pauli limited for $H \,||\, c$. This was suggested for 2-layer systems with the same symmetry configuration [4], but a similar situation emerges for superconductivity in $CeRh_2As_2$ based on the lack of local inversion symmetry at the Ce site [2].

Preliminary results suggest that the normal state part of the $CeRh_2As_2$ phase diagram is also non-trivial. The putative ordered state below $T_0 \approx 0.4$ K seems to be suppressed near $H \approx 4$ T as well, and the corresponding transition may be a fourth one to join the multicritical point. This will be investigated further at our institute.

What is really new in $CeRh_2As_2$ compared to other unconventional superconductors is the possibility of even- and odd-parity superconductivity, even though the formation of Cooper pairs occurs via the same (singlet or triplet) pairing channel for both orders. It is the unusual crystal structure and electronic environment of $CeRh_2As_2$, which open up the possibility of an even to odd parity phase transition by a change of sign of the order parameter in one of the sublayers.

The system presents a number of open questions concerning the mechanism of superconductivity, such as whether its onset in zero applied magnetic field is linked to the preceding unidentified order. That 'hidden' order is probably rooted in the unusual Ce environment as well. In the coming years, we will use $CeRh_2As_2$ as a benchmark material to study the interplay of strong correlations with locally broken inversion symmetry and the subsequent Rashba interaction and their effect on superconductivity and other orders.

External Cooperation Partners

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