

Exposing the odd-parity superconductivity in CeRh₂As₂ with hydrostatic pressure

K. Semeniuk^{1,*}, M. Pfeiffer^{1,2,*}, J. F. Landaeta^{1,2}, M. Nicklas¹, C. Geibel¹,
M. Brando¹, S. Khim¹ and E. Hassinger^{1,2,‡}

¹Max Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany

²Institute for Solid State and Materials Physics, TU Dresden University of Technology, 01062 Dresden, Germany



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Odd-parity superconductivity is a fundamentally interesting but rare state of matter with a potential for applications in topological quantum computing. Crystals with staggered locally noncentrosymmetric structures have been proposed as platforms where a magnetic field can induce a transition between even- and odd-parity superconducting (SC) states. The strongly correlated superconductor CeRh₂As₂, with the critical temperature $T_c \approx 0.4$ K, is likely the first example material showing such a phase transition, which occurs at the magnetic field $\mu_0 H^* = 4$ T applied along the crystallographic c axis. CeRh₂As₂ also undergoes a phase transition of an unknown origin at $T_0 = 0.5$ K. By subjecting CeRh₂As₂ to hydrostatic pressure and mapping the resultant changes to the SC phase diagrams we investigated how the lattice compression and changes to the electronic correlations affect the stability and relative balance of the two SC states. The abnormally high in-plane upper critical field becomes even higher close to a quantum critical point of the T_0 order. Remarkably, the SC phase-switching field H^* is drastically reduced under pressure, dropping to 0.3 T at 2.7 GPa. This result signals an apparent strengthening of the local noncentrosymmetry and forecasts a possible stabilization of the putative odd-parity state down to zero field, hitherto not considered by theoretical models.

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Introduction. Broken inversion symmetry in crystals causes momentum-dependent spin polarization, as exemplified by the Rashba effect [1]. In noncentrosymmetric superconductors, coupling of the momentum and spin degrees of freedom prohibits labeling the Cooper-pair wave function as a purely spin-singlet or spin-triplet one — the state is a mix of both [2]. However, two noncentrosymmetric lattices can be combined into a globally centrosymmetric superstructure, with the constituents related via the inversion symmetry. The superconducting (SC) state must then have a definite parity and a magnetic-field-induced switching from an even-parity state to an odd-parity state is expected when the polarization of the spins due to the sizable spin-orbit coupling remains preserved locally and stabilizes the odd-parity state [3].

The two-phase superconductivity in CeRh₂As₂ may be the only known practical realization of this phenomenon [4]. The material crystallizes in a nonsymmorphic CaBe₂Ge₂-type structure (space group 129, P4/nmm) [5], and in accordance with the above-mentioned principle, its unit cell has two noncentrosymmetric Ce sites of opposite polarity, related to each other via inversion symmetry.

CeRh₂As₂ is an unconventional superconductor with the critical temperature $T_c \approx 0.3 - 0.4$ K, varying with sample quality [4,6]. Applying a magnetic field $H_{||}$ parallel to the crystallographic c axis reveals two peculiar aspects: 1) At $\mu_0 H^* = 4$ T, a transition from a low-field SC state (SC1) to a high-field state (SC2) takes place; 2) The upper critical field $H_{c2}^{||}$ reaches 15 T [4,7], which is unusually high for the given T_c . The SC2 state rapidly vanishes as the field is rotated, and for a field $H_{\perp}$ perpendicular to the c axis, only the SC1 phase exists with the upper critical field $H_{c2}^{\perp}$ of 2 T [7]. Based on a reasonable agreement of these observations with the earlier predictions [3], SC1 and SC2 have been interpreted, respectively, as the states of the even and odd parity of the order parameter [4]. The conjecture is further supported by recent theoretical works [8–10].

At a temperature $T_0 \approx 0.5$ K, CeRh₂As₂ enters a state “Phase I” of currently unknown microscopic origin [6,11,12]. At a pressure $P_0 \approx 0.5$ GPa, Phase I vanishes in a quantum critical point (QCP), which gives rise to the quantum critical fluctuations that apparently drive the SC pairing and lead to the extreme quasiparticle masses and non-Fermi-liquid behavior in the system [4,13]. Additionally, recent nuclear magnetic and quadrupolar resonance studies detected local magnetic fields below 0.25 K and proposed an antiferromagnetic (AFM) order [14–16], but it is currently unclear whether it is concomitant with T_0 or T_c transitions, or exists independently from the other states [17].

The alternating Rashba-type spin-orbit coupling due to the local inversion symmetry breaking and strong electronic correlations of a heavy-fermion system are thought to be the key ingredients for realizing the parity transition. The former favors comparable magnitudes of the SC pairing

*These authors contributed equally to this work.

†Contact author: konstantin.semeniuk@cpfs.mpg.de

‡Contact author: elena.hassinger@tu-dresden.de

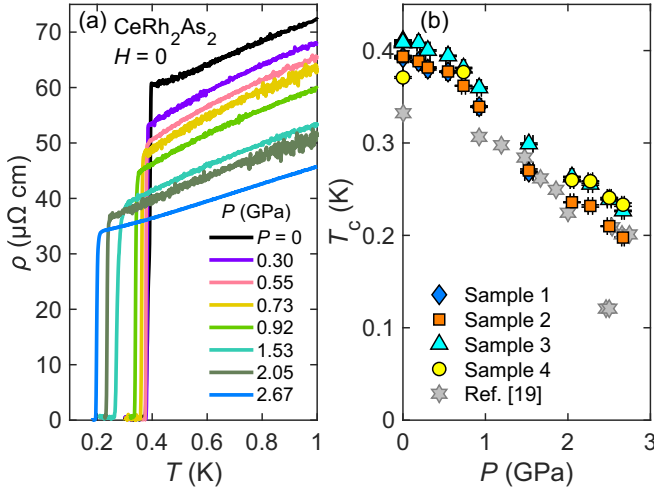


FIG. 1. Effect of pressure on the superconducting transition of CeRh₂As₂. (a) Resistivity (ρ) of CeRh₂As₂ (Sample 2) against temperature (T) at different pressures. (b) Pressure (P) dependence of the transition temperature T_c for different samples, including data from an independent study [19]. T_c is defined as the temperature at which $d\rho/dT$ peaks. Differences between the samples are explained in Supplemental Material [20].

potentials of the two phases. The latter allows the purely orbitally-limited SC2 state to survive at higher fields than the paramagnetically-limited SC1 state. The low orbital limiting field of isostructural weakly-correlated LaRh₂As₂ can then explain why this compound hosts only one SC phase [18].

We used hydrostatic pressure to investigate how tuning both the strength of correlations and the local noncentrosymmetry affects the unusual superconductivity in CeRh₂As₂. While an independent study [19] reported a discontinuity in $T_c(P)$ at 2.5 GPa, hinting at a pressure-induced transition to a different SC state, this result was not reproduced in our parallel work [13], and there are strong indications that the anomaly was caused by a sudden loss of pressure hydrostaticity, rendering the results for $P \geq 2.5$ GPa questionable. The study also suffered from a sizable uncertainty in critical temperatures and fields at high pressures, making it impossible to draw solid conclusions regarding the effect of pressure on the relative balance between SC1 and SC2 (we analyze the results of Ref. [19] in more detail in Supplemental Material [SM]).

In this letter, we demonstrate that the in-plane upper critical field is enhanced and the shape of the $H_{c2}^{\perp}(T)$ curve becomes anomalous close to P_0 , in agreement with the presence of the QCP [13]. More importantly, our study reveals a pronounced and interesting effect of pressure on the multi-phase superconductivity: while both SC1 and SC2 occupy a smaller area in the $H_{\parallel} - T$ space at high pressures, SC2 becomes more favored with respect to SC1 (contrary to Ref. [19]). This is accompanied by a dramatic reduction of the phase-switching field H^* , signifying a major enhancement of the staggered Rashba interaction.

Results. In Fig. 1(a) we show how pressure affects the SC transition seen in $\rho(T)$ at zero magnetic field, and in Fig. 1(b) we plot resistive $T_c(P)$ for multiple samples studied. Besides a ~ 30 mK spread of T_c , all of the measured samples show consistent pressure dependence, which agrees with the one

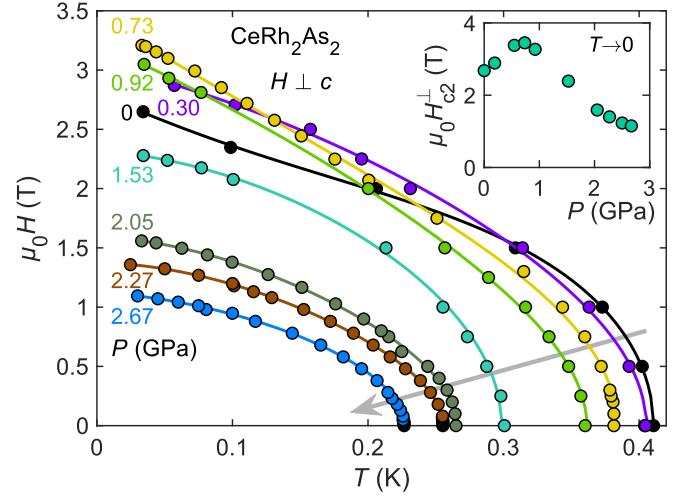


FIG. 2. Superconductivity in CeRh₂As₂ under pressure (P) in the in-plane magnetic field $H \perp c$. (a) Field-temperature ($\mu_0 H - T$) phase diagrams (Sample 3). The solid lines are guides for the eye. The grey arrow indicates the change for increasing pressure. Inset: the zero-temperature upper critical field against pressure.

found in our parallel study [13]. At ambient pressure, the system is close to the peak of the SC dome in the $T - P$ space, and T_c effectively stays at ~ 0.4 K up to about 0.7 GPa. Subsequently, T_c steadily decreases down to ~ 0.2 K at 2.67 GPa, the highest achieved pressure. Compared to the independent high-pressure study of CeRh₂As₂ [19], the overall rate of suppression of T_c is similar, yet the sharp dip near 2.5 GPa is definitely absent for all our samples, indicating that the dip was almost certainly caused by the loss of hydrostaticity [13,20].

The in-plane upper critical field curves, $H_{c2}^{\perp}(T)$, are shown in Fig. 2 for different pressures. A selection of $\rho(T)$ and $\rho(H)$ data behind the displayed phase diagrams is given in SM [20]. For $T \ll T_c$, $H_{c2}^{\perp}$ initially increases with pressure and peaks between 0.55 and 0.73 GPa. At these pressures, the $H_{c2}^{\perp}(T)$ curve follows a rather atypical linear increase in the low-temperature region without signs of saturation down to 30 mK. At higher pressures, $H_{c2}^{\perp}$ decreases in the entire temperature range, and the saturating form expected from a strongly Pauli-limited superconductor is recovered by 1.5 GPa.

The upper critical field curves for the out-of-plane magnetic field, $H_{c2}^{\parallel}(T)$, are depicted in Fig. 3(a). The superconductivity is monotonically suppressed by pressure at all temperatures. In the zero-temperature limit, $H_{c2}^{\parallel}$ is reduced from 18 T to 4 T across the full pressure range [Fig. 3(b)]. The reduction of low-temperature $H_{c2}^{\parallel}$ in the SC2 state mimics the decrease of the orbital limiting field $H_{\text{orb}}^{\parallel}$, estimated from the slope of the critical field curve near T_c , in the SC1 state, but there exists a ~ 4 T offset between the two quantities. The SC1-SC2 transition appears as a pronounced kink in $H_{c2}^{\parallel}(T)$. The kink shifts down in both field and temperature with pressure, and can be tracked all the way to 2.67 GPa, where $\mu_0 H^* = 0.3$ T, indicating that the two SC phases are

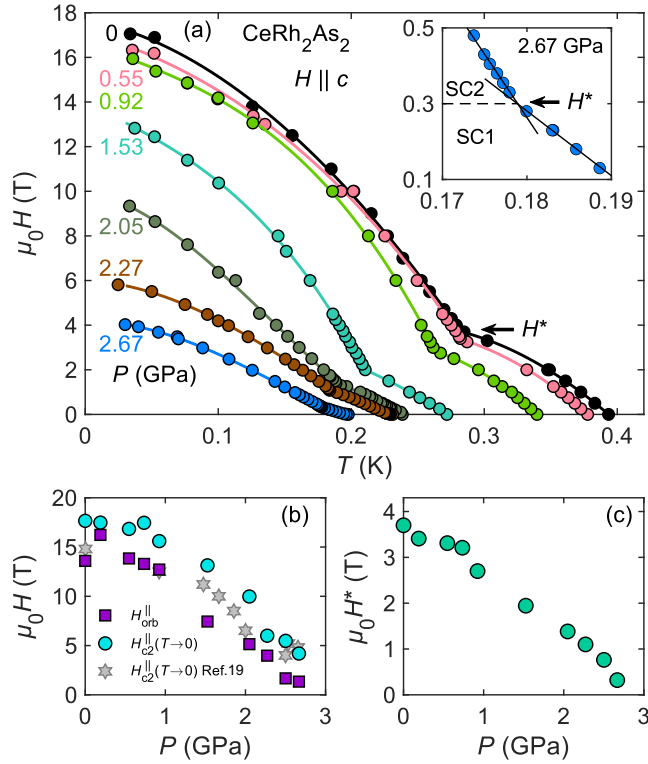


FIG. 3. Superconductivity in CeRh_2As_2 under pressure (P) in the out-of-plane magnetic field $H \parallel c$. (a) Field-temperature ($\mu_0 H$ - T) phase diagrams (Samples 1 and 2). The solid lines are guides for the eye. The transition between the two superconducting phases SC1 and SC2 occurs at a magnetic field H^* and is seen as a kink in the critical field curve (marked for $P = 0$). Inset: the SC1-SC2 transition at 2.67 GPa. The straight trend lines emphasize the slope change. The dashed line marks a hypothetical SC1-SC2 phase boundary. (b) Pressure dependence of the orbital-limiting upper critical field ($H_{\text{orb}}^{\parallel}$), estimated from the slopes of $H_{c2}^{\parallel}(T)$ curves at T_c as well as the zero-temperature limit of the upper critical field $H_{c2}^{\parallel}$. Data from an independent study [19] are included for comparison. (c) Pressure dependence of H^* .

still present [inset of Fig. 3(a)]. Figure 3(c) shows a steady decrease of H^* with pressure.

Discussion. Whereas the anomalously high Pauli limit in CeRh_2As_2 has been mainly attributed to the enhancement caused by the Rashba interaction [4], electronic correlations may also play a crucial role [21]. In particular, strong fluctuations in the vicinity of the Phase I QCP could explain the nontrivial pressure dependence of the SC1 upper critical field for $H \perp c$ and the large Pauli limit. The Chandrasekhar-Clogston limit is violated in a number of other strongly-correlated superconductors, which do not exhibit a pronounced inversion symmetry breaking at the relevant sites [22–25]. In these cases, the large Pauli limit has been attributed to a strong coupling of electrons to the pairing-mediating fluctuations, in analogy to strongly-coupled phonon-mediated superconductors, where an enhancement of the Pauli limit was found to reflect the renormalization of the density of states at the Fermi level [26,27]. The maximum of $H_{c2}^{\perp}$ near P_0 in CeRh_2As_2 resembles the case of other Ce-based heavy-fermion superconductors, such as CeIrSi_3 [28] and

CeCu_2Si_2 [29], where H_{c2} peaks close to the critical pressure of the magnetic order. The atypical shape of $H_{c2}^{\perp}(T)$ can then be explained by a sizable increase of the coupling strength upon approaching the QCP with decreasing temperature.

By multiplicatively scaling the $H_{c2}^{\perp}(T)$ data in Fig. 2 along the temperature and field axes, the critical field curves at different pressures can be shown to follow the same form down to at least $T/T_c \approx 0.5$. This also applies to the ambient-pressure critical field data from the previous heat capacity and magnetic AC susceptibility measurements [4,7]. Note, that in the case of heat capacity and susceptibility, the field has to be scaled by a factor of 2, as opposed to 2.5 for the ambient-pressure resistivity, which reflects the difference in T_c (addressed in SM [20]). This universal behavior can be decently approximated by the theoretical even-parity curve from an earlier study [4] [the dashed curve in Fig. 4(a)], with the agreement even extending to the lowest achieved temperatures at 2.50 and 2.67 GPa. Using the $T/T_c > 0.5$ portion of the $H_{c2}^{\perp}(T)$ data to constrain the theoretical even-parity curve, we define the effective zero-temperature Pauli limit $H_{c2,e}^{\perp}$, which serves as the scaling factor in Fig. 4(a) and does not account for the low-temperature deviation below 2 GPa. The decrease of this Pauli limiting field with pressure can mostly be correlated with the reduction of T_c [Fig. 4(c)].

Close to T_c , the theoretical curve visibly deviates from the data, implying that the initial slope of $H_{c2}^{\perp}(T)$ and hence the in-plane orbital limiting field estimated from it are significantly steeper than found in the earlier experiments [4,7], where the slope was possibly underestimated due to a low density of points. Based on the current data, we place the lower limit of $|(dH_{c2}^{\perp}/dT)_{T_c}|$ at 113 T/K at $P = 0$ [20]. A steep initial slope is indeed expected for superconductivity arising from electronic states with extreme effective mass anisotropy or belonging to quasi-two-dimensional elements of the Fermi surface [11,30,31]. The quasi-two-dimensional Fermi sheets could also explain why the model based on loosely coupled superconducting layers [3] may work for bulk CeRh_2As_2 .

Aside from the influence of the quantum critical fluctuations, the atypical shape of $H_{c2}^{\perp}$ at low temperature could also be caused by a presence of a finite-momentum pairing state [32,33]. A particular variant of it, called a complex stripe phase, has been predicted for systems with locally broken inversion symmetry [34]. The superconductivity in CeRh_2As_2 is strongly Pauli limited for $H \perp c$ and the band structure is likely anisotropic [9,11,30,31] — these features are necessary for enabling a finite-momentum pairing [35]. However, such states are easily destroyed by impurities [35]. We estimate comparable mean free paths l and coherence lengths ξ for the probed batch of CeRh_2As_2 samples (see SM [20]). For the in-plane values we find $l_{\text{ab}} \approx 2\xi_{\text{ab}}$, meaning that the clean limit is possibly applicable for $H \parallel c$. Further studies using bulk probes and newer generations of crystals are needed for investigating the anomalous shape of $H_{c2}^{\perp}$ and the possible existence of the complex stripe phase.

As for the effect of pressure on the multi-phase superconductivity, both SC1 and SC2 persist well beyond the Phase I critical pressure of $P_0 \sim 0.5$ GPa [13]. Besides reinforcing the fact that the SC1-SC2 line is not a transition between a mixed SC+I and a pure SC state [6], this finding also bears consequences for the spin-flop interpretation of the H^*

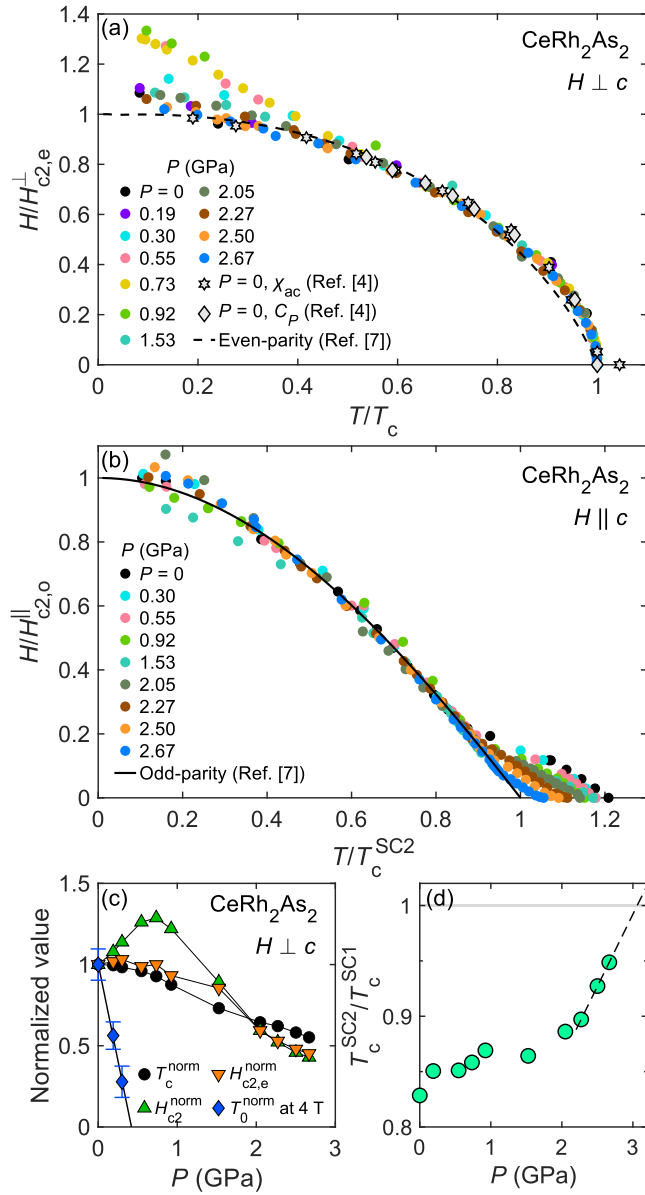


FIG. 4. Summarized effect of pressure (P) on the superconductivity in CeRh_2As_2 . (a) Scaled in-plane critical field curves (T_c –zero-field critical temperature; $H_{c2,e}^\perp$ – effective Pauli-limiting field, based on a theoretical even-parity curve [4,7], shown as the dashed line). Scaled critical field data from the earlier heat capacity (C_p) and AC magnetic susceptibility (χ_{AC}) measurements [4] are included. (b) Scaled out-of-plane critical field curves (T_c^{SC2} –extrapolated zero-field critical temperature of the high-field SC2 phase; $H_{c2,o}^\parallel$ – critical field at $T = 0$ according to a fit to the odd-parity model [4,7], shown as the solid line). (c) Pressure dependence of the relevant properties, normalized to their zero-pressure values, including the critical temperature T_0 of Phase I at 4 T [13] ($H_{c2}^\perp$ – the actual in-plane upper critical field extrapolated to $T = 0$). (d) Pressure dependence of the ratio of the SC2 and SC1 critical temperatures at $H = 0$.

signature, suggested in previous works [16,36]. Namely, if the SC1-SC2 transition involves magnetic ordering, then the latter is entirely separate from Phase I, and is at least as robust under pressure as H^* . Hence, while the T_0 quantum critical transition is a likely source of pairing-mediating interactions, the

phenomenology of the SC1-SC2 transition can be regarded independently of Phase I.

The $H_{c2}^\parallel(T)$ curve of the SC2 state retains the orbitally limited character at all pressures. As shown in Fig. 4(b), by multiplicatively scaling the data for different pressures along the temperature and field axes, the phase boundaries between the SC2 and normal states can be superimposed onto a single theoretical curve for an odd-parity state [7]. Such a universality indicates that changes to the effective quasiparticle mass and T_c are sufficient for explaining the pressure dependence of the SC2 critical field curve. Pressure dependence of the c -axis orbital limiting field indicates that the Fermi velocity increases above 1.5 GPa (see SM [20]), which possibly reflects the weakening of correlations at high pressure observed in the normal-state resistivity [13].

By extrapolating the SC2 critical field curve to $H = 0$, we define a fictive critical temperature T_c^{SC2} and denote the usual T_c as T_c^{SC1} . The ratio $T_c^{\text{SC2}}/T_c^{\text{SC1}}$ increases with pressure and steeply approaches unity for $P > 2$ GPa [Fig. 4(d)], in agreement with H^* decreasing towards zero [Fig. 3(c)].

Theories describing locally non-centrosymmetric superconductors quantify the extent of local inversion asymmetry with a parameter α/t_c , where α is the strength of Rashba interaction and t_c is the interlayer hopping parameter. According to the parity-switching picture, the observed enhancement of T_c^{SC2} relative to T_c^{SC1} implies an increase of α/t_c under pressure, whereas one would naively expect t_c to become larger as the c -axis lattice parameter shrinks, and thus α/t_c to decrease with pressure. The increase of α/t_c could be linked to the prediction of the Fermi sheets relevant for the superconductivity being centered at the edges of the Brillouin zone (BZ), where the nonsymmorphic symmetry of the lattice enforces $t_c = 0$ [9,11]. If these Fermi sheets shift closer to the BZ boundaries under pressure, the mean value of t_c can indeed decrease. Note that the increase of α/t_c should lead to a further enhancement of the Pauli limiting field of SC1. For $H \parallel c$, the effect should be particularly strong, but the SC1 section of the phase diagram becomes too small to investigate that. For $H \perp c$, however, the Pauli limiting field is expected to increase only by a minor amount (discussed in SM [20]) in line with our results.

What is more striking, is the unexpectedly steep increase of $T_c^{\text{SC2}}/T_c^{\text{SC1}}$ above 2 GPa. The trend predicts that above 3 GPa the SC2 phase will become the only stable SC phase, in agreement with the pressure dependence of H^* . According to the model, when the staggered Rashba interaction is present, the ratio of the magnitudes of the odd- and even-parity state order parameters is given by $(\alpha/t_c)/\sqrt{(\alpha/t_c)^2 + 1}$, implying that T_c^{SC2} is always smaller than T_c^{SC1} . This inference, however, assumes the equal bare pairing potentials of the two states (before the Rashba interaction is considered) [9,10]. The strong changes of $T_c^{\text{SC2}}/T_c^{\text{SC1}}$ (and H^*) at high pressure challenge this assumption and provide a new input for extended theories.

Conclusions. Our study of the superconductivity in CeRh_2As_2 under pressure demonstrates a strong overall sensitivity of SC properties to the proximity to the quantum critical point. However, the pronounced stabilization of the SC2 state over SC1 under pressure indicates that a different tuning

parameter drives the balance between the two SC phases. The proposed relevance of the staggered Rashba interaction fits this picture and implies a major increase of the local non-centrosymmetry with pressure. These results demonstrate a good tunability of the system and motivate further experimental and theoretical investigations into the microscopic processes involved in the superconductivity and other ordered states of CeRh_2As_2 .

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