Discovery of Magnetic Phase Transitions in Heavy-Fermion Superconductor $CeRh_2As_2$

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We report on the specific heat studies performed on a new generation of $CeRh_2As_2$ single crystals. Superior quality of the samples and dedicated experimental protocol allowed us to observe an antiferromagneticlike behavior in the normal state and to detect the first-order phase transition of magnetic origin within the superconducting state of the compound. Although in the available literature the physical behavior of $CeRh_2As_2$ is most often described with the use of quadrupole density wave scenario, we propose an alternative explanation using analogies to antiferromagnetic heavy-fermion superconductors $CeRhIn_5$ and Ce_2RhIn_8.

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Heavy-fermion superconductivity is one of the most intriguing phenomena of modern solid-state physics. Its full understanding seems to remain elusive at the current stage, and continuous studies bring new questions and challenges. The recently discovered heavy-fermion superconductor $CeRh_2As_2$ [1] focused enormous attention of solid-state community due to its unusual features. One of these properties is the phase transition within the superconducting state, which occurs upon applying magnetic field along the c axis and leads to an extraordinary violation of the Pauli-Clogston limit. It also makes $CeRh_2As_2$ one of the only few known multiphase heavy-fermion superconductors (together with, e.g., UTe_2 [2,3] and UPt_3 [4–6]). The second interesting feature of the compound is the enigmatic T_0 anomaly, observed at temperature slightly above the superconducting phase transition. Although numerous experimental investigations [7–15] supported by various theoretical studies [16–24] have been performed up to date, there is still no decisive explanation for the nature of both these peculiarities. It has been proposed that $CeRh_2As_2$ hosts a quadrupole density wave (QDW) instability [7] and the anomaly at T_0 is a signature of the entrance to this unique nonmagnetic ordered state. Interestingly, Machida recently proposed [24] the antiferromagnetic (AFM) ordering picture for an explanation of the anomaly in the field dependence of magnetic susceptibility, $\chi(H)$, of $CeRh_2As_2$, which marks the transition between its two superconducting states. Here, we show that the same scenario can also describe the behavior of the specific heat data, $C_p(T)$, of this compound without including the QDW concept.

The insufficient homogeneity of the samples reported so far, responsible for significant broadening of thermodynamic features related to the phase transitions, is an essential problem in the proper understanding of the physical behavior of $CeRh_2As_2$. In general, high-quality samples are crucial for detailed investigation of all low-temperature unconventional superconductors. As recent studies on UTe_2 [25,26], Sr_2RuO_4 [27,28], and $YbRh_2Si_2$ [29,30] have shown, the superior quality of samples, either solely, or combined with other developments (like strain tuning of the critical temperature or precise temperature control of the experimental setup) provided important insights into the fascinating nature of these materials. Recently Semeniuk *et al.* reported on a new batch of single crystals of $CeRh_2As_2$ [15], the quality of which was claimed to be the highest among those investigated before, based on a few parameters derived from the thermodynamic and electrical transport data. Analysis of the very same set of parameters for crystals synthesized and studied in the present work (see Supplemental Material [31]) indicated that they are of even higher quality. The already reported anomalies in $CeRh_2As_2$ have been reproduced in our measurements, however, they are sharper and clearer. Most importantly, we have also observed a new feature in $C_p(T)$, which was not found in the previous studies possibly because of inhomogeneity of samples and/or different experimental protocols.

Figure 1 presents low-temperature $C_p(T)$ data collected in several different magnetic fields applied along the c axis for the new generation of $CeRh_2As_2$ single crystals. As can be easily noticed, the zero-field curve exhibits two distinct anomalies. A smaller one, at $T_0 = 0.50$ K, has been proposed to be the manifestation of a QDW [7,15]. In turn, the larger one, at $T_{sc} = 0.32$ K, is related to the superconducting phase transition. The determined temperatures are distinctly higher than temperatures originally reported by Khim *et al.* [1] and comparable with the ones revealed recently [15]. Above T_0, up to about $T = 3$ K for the zero-field data (for the extended data range see Supplemental Material [31]), the normal state $C_p(T)$ curve can be very well described by logarithmic temperature dependence $A\ln(T/T^*)$, where A and T^* are parameters of the fit. The same relation can be also fitted at least up to 1 K to the corresponding experimental data acquired with

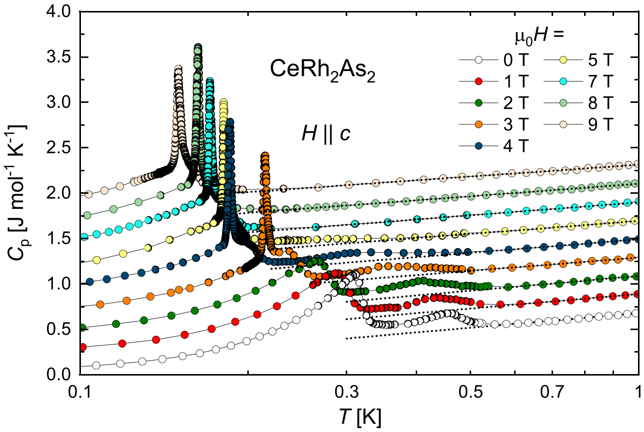

FIG. 1. Specific heat of $CeRh_2As_2$ in magnetic fields applied along the c axis. Straight dashed lines emphasize a logarithmic behavior. For clarity, the consecutive curves were shifted upward by 0.2 $J\,mol^{-1}\,K^{-1}$.

applied magnetic fields [see the straight dashed lines in Fig. 1]. While the parameter A with increasing magnetic field changes only slightly (from 0.26 $J\,mol^{-1}\,K^{-1}$ for 0 T to 0.21 $J\,mol^{-1}\,K^{-1}$ for 9 T), the corresponding change of T^* is much more pronounced (from 53 to 23 mK). It should also be noted that with the shift of T_0 toward lower temperatures, the low-temperature limit of the applicability of this relation lowers accordingly. Assuming that the phonon contribution to the specific heat of $CeRh_2As_2$ is similar to that determined for its nonmagnetic counterpart $LaRh_2As_2$ ($\beta = 0.34$ $mJ\,mol^{-1}\,K^{-4}$ [36]), it can be safely stated that below 1 K the lattice contribution to C_p is negligibly small compared to the measured specific heat in this temperature range. Hence, the logarithmic term $\sim \ln T$ provides a good description of the sum of electronic and magnetic contributions to $C_p(T)$ of $CeRh_2As_2$. This is somehow surprising, since in Ce-based compounds for the low-temperature non-Fermi-liquid behavior one usually expects the $\sim T \ln T$ dependence of the specific heat. Interestingly, based on the high-pressure electronic specific heat data of Ce_2RhIn_8 [37] we found that, regardless of the applied pressure, the high-temperature tail of the antiferromagnetic anomaly in Ce_2RhIn_8 in the temperature range from about 3 to 7 K can be very well described by the same logarithmic function $A \ln(T/T^*)$, as was used to describe the normal state specific heat of $CeRh_2As_2$.

As can be seen in Fig. 1, with increasing strength of the magnetic field, T_0 anomaly broadens and systematically shifts toward lower temperatures, which is a feature typical for antiferromagnets. It can also be noticed that because with increasing magnetic field the anomaly at T_{sc} shifts toward lower temperatures slower compared to the T_0 anomaly, with rising field strength they get closer and closer to one another and above about 5 T only a single distinct anomaly is visible.

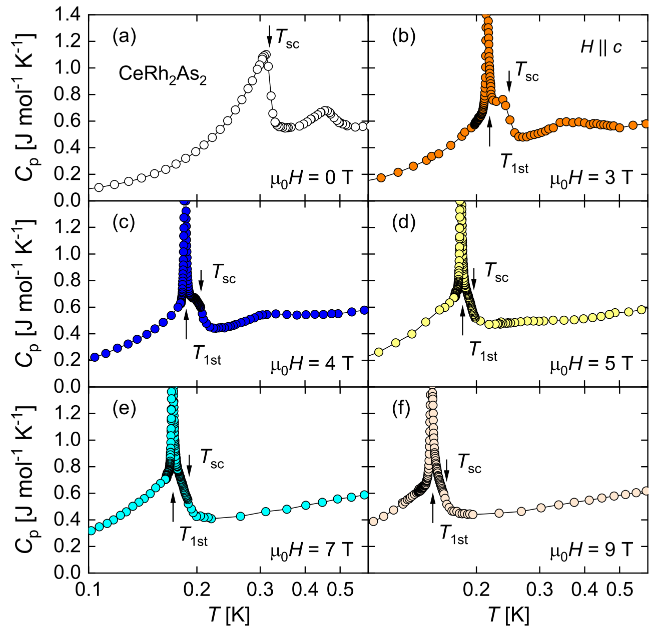

FIG. 2. Evolution of the anomalies in the specific heat of $CeRh_2As_2$ in magnetic fields of (a) 0 T, (b) 3 T, (c) 4 T, (d) 5 T, (e) 7 T, and (f) 9 T applied along the c axis. The arrows mark the phase transitions at T_{sc} and T_{1st}.

Importantly, by performing specific heat measurements with the use of the long heat pulse method [34], in magnetic fields higher than about 3 T, we were also able to detect a first-order phase transition which occurs within the superconducting state of the compound. No such transition was clearly visible in $C_p(T)$ curves either for zero magnetic field [see Fig. 2(a)] or any other magnetic field smaller than about 3 T. Taking a closer look at the specific heat curves measured in different magnetic fields, one can observe that in 3 T the superconducting phase transition at T_{sc} and the first-order transition at T_{1st} are clearly separated, as shown in Fig. 2(b). In higher fields they approach each other [see Fig. 2(c)] and above about 5 T [Fig. 2(d)] the first-order-type anomaly follows the superconducting one as if they are coupled, shifting together toward lower temperatures with a further increase of magnetic field [Figs. 2(e) and 2(f)]. Because to the best of our knowledge no such behavior has been reported to date for a purely superconducting system, we suppose that the first-order transition is closely related to the T_0 anomaly. In turn, very similar field-induced phase transitions have already been observed, e.g., for antiferromagnetic superconductors $CeRhIn_5$ and Ce_2RhIn_8 [35,38]. For both compounds, the first-order phase transition has been proven to occur due to the reconstruction of their antiferromagnetic structures [39,40]. Therefore, the presence of the phase transition at T_{1st} can be considered as a strong support of the AFM scenario, which presumes that the T_0 anomaly in $CeRh_2As_2$ is a manifestation of some kind of long-range magnetic ordering. Such previously unobserved behavior

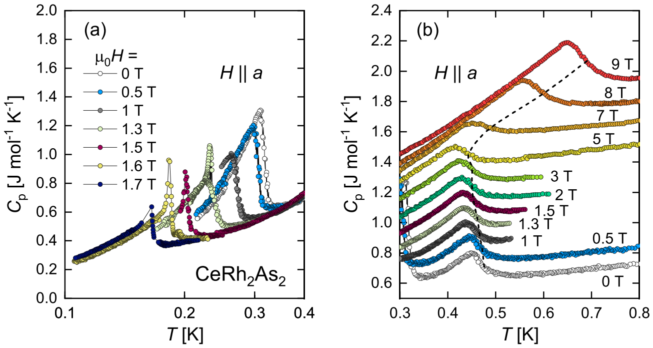

FIG. 3. Specific heat of $CeRh_2As_2$ near the (a) superconducting transition and (b) magnetic transition in magnetic fields applied along the a axis. The dashed curve tracks the position of T_0. For clarity, the consecutive curves were shifted upward by 0.1 J mol^{-1} K^{-1}.

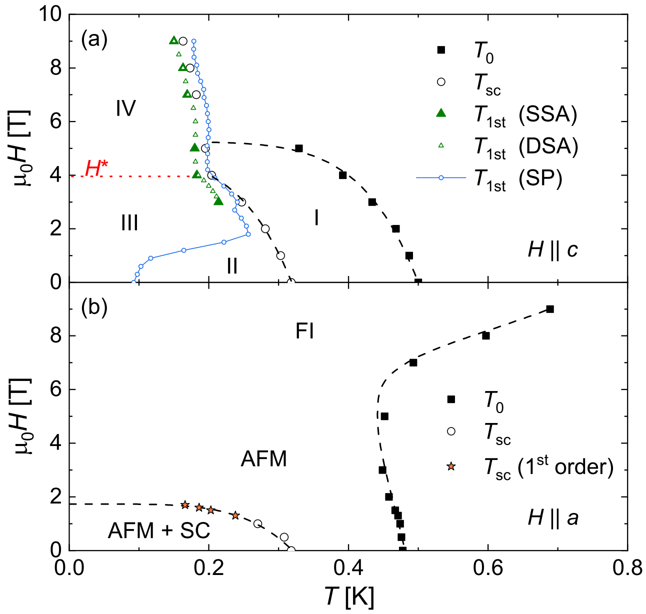

FIG. 4. H-T phase diagrams of $CeRh_2As_2$ constructed from the specific heat data collected in magnetic field applied along (a) the c axis and (b) the a axis. Dashed lines are guides for the eye. The red dotted line marks the approximate position of the H^* anomaly taken from the magnetic susceptibility data [1]. For the explanation of abbreviations see the main text.

may also provide an alternative explanation of the mechanisms governing the change of the superconducting state of $CeRh_2As_2$, to be discussed later. More precise studies of the evolution of the first-order phase transition were performed using a set of various specific heat measurement approaches, and the details are described in Supplemental Material [31].

Figure 3 displays low-temperature specific heat data of $CeRh_2As_2$ collected for $H \| a$. As can be inferred from Fig. 3(a), in this configuration, the behavior of the superconductivity-related anomaly is initially typical for most superconductors. With increasing magnetic field, the anomaly at T_{sc} shifts to lower temperatures and its magnitude diminishes. Interestingly, in a field of about 1.3 T, the peak in $C_p(T)$ becomes distinctly sharper, hence suggesting a change in character of the phase transition from the second to first order. A similar effect was observed in, e.g., $CeCoIn_5$ [41] and Ce_2PdIn_8 [42] and was ascribed to the possible emergence of low-temperature high-field FFLO (Fulde-Ferrell-Larkin-Ovchinnikov) state [41,43]. It is a tempting conjecture that the observed behavior is a manifestation of FFLO state formation in $CeRh_2As_2$, however, this hypothesis needs solid experimental verification.

The evolution of the T_0 anomaly in $H \| a$ is presented in Fig. 3(b). Since measurements for this field direction were performed on a different piece of single crystal, one can notice slight differences in the shape of the T_0 anomaly. Compared to that used for $H \| c$ measurements, it is sharper and a little higher, which suggests even better homogeneity of this part of the crystal. As a consequence of reduced broadening of the anomaly, the transition temperature $T_0 = 0.48$ K is somewhat lower. As can be noticed, for fields lower than about 5 T, with increasing field strength T_0 slightly decreases, as expected for antiferromagnets. Above 5 T, this trend changes and further increase of the magnetic field results in a distinct shift of the anomaly toward higher temperatures. At the same time the λ-shaped anomaly becomes sharper. This strange feature of $CeRh_2As_2$ has not been reported in the previous studies.

Based on our specific heat data, we constructed the phase diagrams of $CeRh_2As_2$, which are presented in Fig. 4. T_{sc} and T_0 were determined by equal entropy construction. In turn, the values of T_{1st} for data obtained by both single-slope analysis (SSA) and dual-slope analysis (DSA) (details on both analysis methods can be found in, e.g., Ref. [44]) were taken from the positions of the maxima of the anomalies. In the diagram for $H \| c$, we also included the field variation of T_{1st} measured by the sample platform (SP) thermometer for a base temperature of 60 mK, here denoted as T_{1st} (SP) (see Supplemental Material [31] for details). It should be noted that values of T_{1st} that are derived in different ways are in very good agreement with each other and a little shift of the T_{1st} (SP) line toward higher temperatures results from the different definition of the transition temperature used in this case. The overall shape of the diagrams, except for the T_{1st} line, that is shown here for the first time, is consistent with the previously reported ones [1,14,15]. The newly discovered T_{1st} boundary appears only for $H \| c$, and in small magnetic fields exists well below T_{sc}. However, with increasing magnetic field strength its position gets closer to T_{sc}, still staying within the superconducting state. In $H \approx 4$ T (being very close to H^* determined from the magnetic susceptibility measurements [1]), the anomalies at T_{sc} and T_{1st} merge, and in higher fields, they follow the same path.

In view of the presented experimental results, we describe $CeRh_2As_2$ as an antiferromagnetic superconductor, which in magnetic fields applied along the c axis undergoes metamagneticlike phase transition at T_{1st}, related to some change in the spin structure. Within this scenario, several different regions of the phase diagram of $CeRh_2As_2$ can be distinguished for $H\|c$ [see Fig. 4(a)]. In region I, the compound is likely an antiferromagnet with the magnetic moments aligned most probably along the c axis. One should note that this picture agrees with one of the proposed magnetic structures describing the AFM state detected by the nuclear quadrupole resonance (NQR) measurements [11]. However, by analogy to Ce_mRhIn_{3m+2} compounds, one can expect for $CeRh_2As_2$ a more complex magnetic structure (such as incommensurate spin density wave) rather than simple AFM phase. At lower temperatures (region II), antiferromagnetism and superconductivity coexist in the material, similarly as was observed for $CeRhIn_5$ and Ce_2RhIn_8 under external pressure [45–49]. The transition from region II to region III may signal the transformation of the magnetic structure of $CeRh_2As_2$, possibly into a commensurate colinear AFM one. Consequently, in region III, rising magnetic field brings about gradual canting of the moments initially oriented antiparallel to the field direction. In a field H^*, a metamagnetic transition occurs, possibly of a spin-flop character, and in fields $H > H^*$ (region IV), a growing ferromagnetic component develops. Thus, in this region the superconducting state coexists with the field-induced ferromagneticlike order instead of the AFM one. As a support for this hypothesis, one may consider the S shape of the temperature dependence of $H_{c2}(T)$, determined for the high-field superconducting phase, that resembles those observed for ferromagnetic superconductors UGe_2 and UCoGe (see, e.g., Ref. [50]). Moreover, our scenario is compatible with the theoretical description proposed recently by Machida [24].

Although the aforementioned NQR experiments [11], as well as recent nuclear magnetic resonance (NMR) studies [51] detected the antiferromagnetic ordering in $CeRh_2As_2$, it needs to be emphasized that according to those studies, the AFM order emerges within the low-field superconducting state of the compound, i.e., well below the T_0 temperature. This finding clearly differs from our interpretation of the T_0 as the temperature of transition to the ordered magnetic state. However, we also notice some similarity. In our scenario, for $3\ \mathrm{T} < H < H^*$, the commensurate AFM state seems to appear at a temperature very close to that of the emergence of the AFM order in the NQR and NMR studies. For lower fields, due to different measurement conditions [in NQR and NMR measurements external magnetic field and temperature are kept stable, while in our measurement of T_{1st} (SP) only the magnetic field is static, but temperature changes dynamically], it is hard to reliably compare our results to those reported before. Nevertheless, the most striking difference is observed for $H > H^*$. In this region, our results show that T_{1st} merges with T_{sc}, hence magnetic ordering appears in the material just after (or together with) the superconductivity emergence. Contrarily, according to the results of NMR measurements [51], antiferromagnetism is no longer observed upon transition to the high-field superconducting phase. This leads to the question, what is the origin of T_{1st} when $H > H^*$, if no magnetism is present in the material in this region, as suggested by NMR studies.

The phase diagram constructed for $H\|a$ is much simpler [see Fig. 4(b)]. It includes the superconducting phase (SC) which exists within the AFM state and two regions of magnetism of a different character. The antiferromagneticlike behavior is observed in fields up to about 6 T. In higher fields, most probably due to a field-induced spin structure reconstruction (FI region), the anomaly in $C_p(T)$ shifts toward higher temperatures, typically for ferromagnets. This behavior distinctly differs from that observed for $H\|c$. However, one should notice that in our scenario, when a magnetic field weaker than about 6 T is applied along the a axis, the orientation of magnetic moments with respect to the external magnetic field might resemble their orientation above spin-flop transition (above 4 T) while the magnetic field is applied along the c axis. In both these cases, the transition temperature slowly shifts toward lower temperatures with increasing field strength. Hence, it is possible that for $H\|c$, the field used in our experiments was not strong enough to polarize magnetic moments into a ferromagneticlike state. One should also take into account the anisotropy of the system as a possible reason for this behavior. To clarify the actual nature of the magnetism and spin structures in $CeRh_2As_2$, further meticulous investigation is necessary.

Based on our interpretation of the collected specific heat data, the T_0 anomaly is ascribed to AFM ordering, despite no distinct signature of the phase transition at T_0 being found so far in the magnetic susceptibility data [1]. This finding resembles, e.g., the case of pressurized $EuTe_2$ [52], where in pressures higher than about 10 GPa AFM order manifests itself in $C_p(T)$ as a clear λ-shaped peak at T_N, but there is no related anomaly in $\chi(T)$. Another explanation of the lack of any clear feature in the magnetic data at T_0 may refer to insufficient quality of the first generation of $CeRh_2As_2$ single crystals [1], for which even the specific heat anomaly at T_0 was hardly visible and the anomaly in $\chi(T)$ could possibly be overlooked. Perhaps, a clear magnetic fingerprint of AFM order could be detected in the newly grown samples. Unfortunately, we are not capable of measuring magnetic susceptibility on our single crystals at such low temperatures in order to verify this hypothesis.

Another issue is related to the magnetic entropy released by T_0 in $CeRh_2As_2$, which for our single crystals equals about 12% of $R\ln 2$. It is only a small fraction of the value expected for the doublet ground state. However, for example, in pressurized AFM superconductor Ce_2RhIn_8, a similar reduction of entropy at T_N ($0.1R\ln 2$ in $p \approx 1.65$ GPa), accompanied with a strong suppression of the peak in $C_p(T)$, was observed [35].

Our detailed specific heat studies of newly grown $CeRh_2As_2$ single crystals allowed us to accurately trace the magnetic field evolution of the T_0 anomaly, which led us to a conclusion on its antiferromagneticlike character. To describe the magnetically ordered state, we used the scenario proposed recently for the explanation of the magnetic susceptibility features of the compound [24]. We suggested that the AFM order in $CeRh_2As_2$ may appear above the onset of superconductivity, i.e., as it happens in every other known magnetic heavy-fermion superconductor. This scenario allowed us to consistently describe all our experimental data without resorting to the concept of QDW formation at T_0. It should be noted that the latter picture implies that $CeRh_2As_2$ showing the onset of AFM inside the superconducting dome would be unique amid heavy-fermion superconductors. We also pointed out a few similarities of $CeRh_2As_2$ to heavy-fermion antiferromagnetic superconductors $CeRhIn_5$ and Ce_2RhIn_8. Moreover, due to the high quality of our single crystals, we revealed the first-order phase transition at T_{1st}, which was not detected in any previous investigation. We ascribed it to the transformation of the magnetic structure of the compound, tentatively from incommensurate spin density wave to the commensurate linear AFM one. In our opinion, another change in the magnetic structure, observed in $H^* \approx 4$ T, may play a role of a driving force for the change of the superconducting state of $CeRh_2As_2$.

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