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2005 J. Phys.: Condens. Matter 17 L231

(http://iopscience.iop.org/0953-8984/17/23/L02)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **17** (2005) L231–L234 doi:10.1088/0953-8984/17/23/L02

LETTER TO THE EDITOR

Novel superconductivity coexisting with incipient electric-multipolar order in $CePt_3Si$

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Received 3 March 2005, in final form 1 April 2005
Published 27 May 2005
Online at stacks.iop.org/JPhysCM/17/L231

Abstract
We measured the specific heat C_p of an annealed sample of $CePt_3Si$ in magnetic fields B up to 10 T and confirmed two conspicuous anomalies in the temperature range where no apparent magnetic anomalies were found in the susceptibility. The lower-temperature anomaly at 2.2 K was depressed with increasing B and seemed to get absorbed around 4 T into the other one around 2.8 K. The latter anomaly continues to grow into a solid peak with increasing B. These features imply that neither of these transitions is due to long-range antiferromagnetism (AF) as originally proposed, but they are reminiscent of quadrupolar transitions found in certain Ce-based Kondo compounds, in which two distinct anomalies were also observed in C_p and identified as a quadrupolar transition followed by an AF transition at a slightly lower T. In the case of $CePt_3Si$, the lower-T anomaly is not due to AF but to a new phase transition of yet-unknown nature, and in this new phase the superconductivity emerges at 0.8 K. We thereby hypothesize that the superconductivity could be mediated by magnetic or other electronic excitations under the influence of the quadrupolar-like order induced around 2.8 K and that $CePt_3Si$ would be the first example of an unconventional superconductor coexisting with electric-multipolar order.

(Some figures in this article are in colour only in the electronic version)

Strongly correlated electron systems have provided a vast fascinating new field to low-temperature physicists. In particular, a recent discovery of unconventional superconductors of various kinds in so-called f-electron heavy-fermion compounds is even amazing [1]. Magnetically mediated pairing, valence-fluctuation-mediated pairing, density-fluctuation-mediated pairing and other forms of pairing have been proposed for such superconductors. Depending upon the magnitude of the exchange interaction parameter J_{cf} between conduction electrons and f-electrons, an electronic instability in the degree of freedom of spin or charge is induced and different pairing interactions are believed to arise in these materials, even under high pressure in some materials. $CePd_2Si_2$, $CeIn_3$, $CeRh_2Si_2$, $CeCu_2Si_2$ and Upt_3 are such examples [2–4]. It is $CePt_3Si$ that has most recently been added to the list of these superconductors [5]. The latter is claimed as a new superconductor without an inversion symmetry, and its pairing mechanism is actively pursued at the present time [6, 7].

We recently reported together with other basic properties that an annealed sample of $CePt_3Si$ exhibits nonmagnetic heavy-fermion behaviour, although a quenched sample does undergo an AF transformation as originally proposed by Bauer *et al* [5] and the indisputable anomaly disclosed by the specific heat at around 2.2 K was tentatively conjectured as a sort of quadrupolar transition rather than an AF one [8]. If this were the case, the superconductivity appearing below 0.8 K ($=T_C$) coexists with the quadrupolar order. In our previous report, however, C_p measurements in external magnetic fields B were carried out only on our old quenched sample and the results were not necessarily very convincing [8]. In order to reveal the more precise nature of the 2.2 K transition, another C_p measurement, this time using a relaxation method, was performed above about 0.6 K in fields B up to 10 T on a small piece of 0.72 mg cut from the same batch as the previously characterized annealed sample [8]. We also verified that there is no anomaly implying a magnetic phase transition down to 0.48 K with a new SQUID magnetometer annexed to a QD magnetometer[5]. The magnetization curve at 1.9 K shown in figure 1 of [8] corroborates that no new magnetic order develops below 2.2 K. These facts are consistent with the specific-heat results shown in the following.

At this point we would like to make a brief comment on other recently published works on $CePt_3Si$ [7, 9], which seem to be contradictory with our findings presented here. We have disclosed that the antiferromagnetism found in our quenched samples was not a bulk property of this compound, and that it disappeared by annealing, and we suggested that it might be caused by strain due to quenching [8]. Taking these facts into consideration, henceforth, we must more carefully examine whether samples under investigation are uniform and really in a thermal equilibrium state or not.

The C_p/T versus $\ln T$ plot is shown for several magnetic fields in figure 1. We first notice a small but distinct anomaly at 2.2 K and a sort of shoulder around 2.8 K on the zero-field curve (denoted by circles), tentatively denoted as T_{Q2} and T_{Q1}, respectively. It is important to note further that the former anomaly is rapidly depressed and absorbed by the latter below about 4 T, and that the latter anomaly becomes sharper and grows into a distinct peak as the field increases towards 10 T, hinting at a more complete phase transition in a higher field, as can be seen a little more clearly in the C_p versus T plot of figure 2. This behaviour of C_p growing with increasing B is not very usual, and is reminiscent of that known as the quadrupolar transition at 3.3 K in CeB_6 or at 1.2 K in $Ce_3Pd_{20}Ge_6$ [10–12]. It would be reasonable to assume that the physics behind these two transitions seems to be very similar, although the transition, which was observed for single-crystalline samples, was much sharper for the latter compounds. We hereby conjecture by analogy to the case of CeB_6 that the transition at T_{Q1} would be an incipient quadrupolar or a more general multipolar transition. However, with respect to the behaviour below these transitions there is a very important difference between them: in CeB_6 an AF transition occurs at 2.5 K in the quadrupolar-ordered phase, while the transition at $T_{Q2} = 2.2$ K in the present compound is not AF but something else of yet-unknown character but somehow related to the transition at T_{Q1}. Anyhow, below these temperatures the superconducting state emerges and coexists with the new ordered state. It is interesting to note further that the superconductivity is destroyed by a similar field around 4 T as the peak at T_{Q2}

[5] The susceptibility below 2 K was measured with a recently developed ^{3}He refrigerator annexed to a QD MPMS, which is now available from iQUANTUM. For more information, contact IQUANTUM Corporation (http://www.iquantum.jp) at info@iquantum.jp.

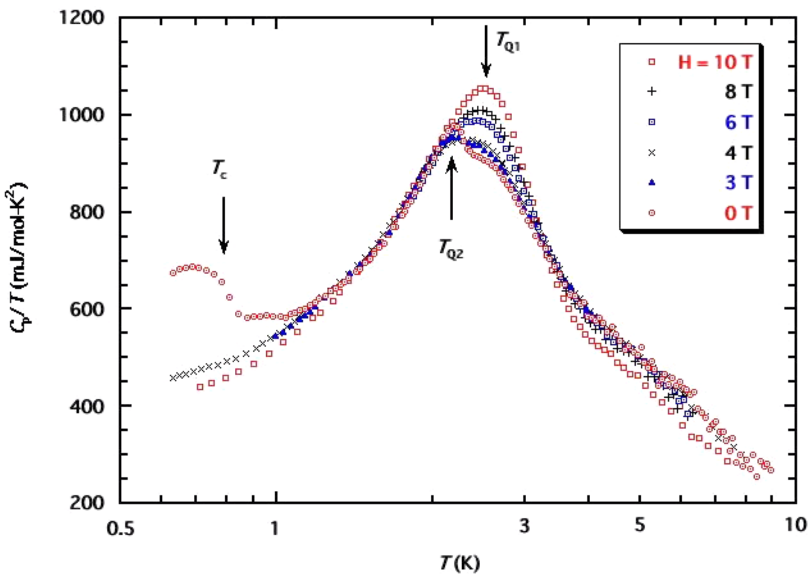

Figure 1. C_p/T versus ln T as a function of several magnetic fields for an annealed sample of $CePt_3Si$. T_{Q1}, T_{Q2} and T_c are respectively two quadrupolar-like transitions and superconducting transition temperature, as described in the text.

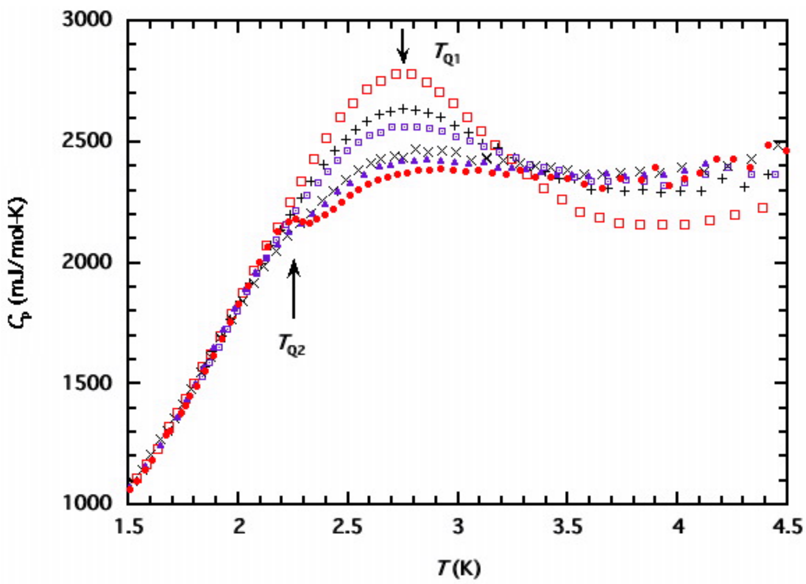

Figure 2. C_p versus T plot for $T = 1.5$–4.5 K. The symbols are the same as in figure 1.

is smeared out, as can be seen in figure 1. This may be just a coincidence, but it may imply that the new phase appearing at T_{Q2} somehow assists the appearance of the superconducting phase. Finally we make a few comments on the C_p curve around T_c. The superconducting transition itself is not as sharp as the one for the bulk sample reported before [8], probably because of it being a tiny sample chipped off a bulk sample. On the other hand, the small anomaly just above T_c has also been seen for the bulk sample previously measured, but only for annealed samples [8]. The anomaly seems to be intrinsic but its nature is not yet known. Characterization of the superconducting phase of the annealed sample, including this point, is now in progress and results will be published elsewhere soon.

We now briefly consider why such a type of coexistence could be feasible in the present compound. One should first realize that the heavy-fermion state with a relatively large exchange interaction parameter J_{cf} is formed closer to an intermediate-valence region rather than a quantum critical point QCP on the verge of an AF phase. A large J_{cf} and high Kondo temperature are inferred from the exceedingly low magnetic entropy at these low temperatures, which is smaller than half of that expected for the two lowest doublets[6]. In other words, it is in a state where strong fluctuations of any kind are conceivable near the presumed multipolar transition, and the superconductivity could be mediated via such fluctuations of spin, valence or charge, as proposed for superconductivity in other heavy-fermion compounds like $CePd_2Si_2$ and $CeCu_2Si_2$ [2–4]. It is therefore vitally important to decide which type of fluctuation is most effective for the superconductivity in $CePt_3Si$.

We thank Dr N Shirakawa for the supplementary susceptibility measurement down to 0.5 K with a SQUID magnetometer in a ^{3}He refrigerator.

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[6] In [8] an overestimated relative value by a factor two for the entropy has erroneously been given.