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LETTER TO THE EDITOR

Ferromagnetic interaction and superconductivity of $CeCu_2Si_2$

M Ishikawa, N Takeda, P Ahmet[1], Y Karaki and H Ishimoto

Institute for Solid State Physics, University of Tokyo, Kashiwanoha 5-1-5, Kashiwa,
Chiba 277-8581, Japan

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Abstract

We have carefully reinvestigated the superconducting heavy-electron system $CeCu_2Si_2$ by preparing very homogeneous polycrystalline samples at many different compositions in the homogeneity range by levitation melting and discovered in the Cu-deficient region a weakly ferromagnetic phase, presumably due to heavy quasiparticles of the Kondo compound. We, in addition, confirmed that the superconducting phase emerges at the fading end of the novel ferromagnetic phase in a very limited region of the phase diagram, which has revealed some salient superconducting properties.

We have carefully characterized many samples of the superconducting heavy-fermion system $CeCu_2Si_2$ [1], mainly by measuring the specific heat, electrical resistivity and magnetic susceptibility. The samples were prepared at about 20 different compositions in the homogeneity range of the compound [2] (see figure 1), by levitation melting in a high-frequency induction furnace, and annealed in an evacuated quartz tube at 850 °C for ten days. The annealed samples were examined by x-ray diffraction (Cu Kα) and metallography, and found to be very homogeneous, as also indicated by their physical properties which will be presented below. Some samples showed a single weak impurity peak at $2\theta = 33^\circ$, which may be the strongest peak of Ce_2CuSi_3 or $CeSi_2$, but the weak peak disappeared after reannealing at 750 °C for six days. The details of the preparation will be published elsewhere soon [3]. The specific heat was measured between 0.15 and 25 K by a quasiadiabatic heat pulse method in a ^{3}He cryostat and a dilution refrigerator, the magnetic susceptibility was measured with a commercial SQUID magnetometer in 3 kOe and the electrical resistivity was measured between 0.03 and 300 K with a dc four-terminal method.

Figure 2 shows (a) the low-temperature susceptibility and (b) M versus H curves taken at 2 K for several samples along the Ce 20.25% line. We can see from these results that the samples No 68, No 61 and No 62 in the Cu-deficient region are weakly ferromagnetic below about 12 K. As the Cu content is increased, the ferromagnetism becomes weaker and the samples are paramagnetic beyond a Cu content between those of No 62 and No 67. Similar variation of magnetic behaviour was also found along the Ce 20% line, where only

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[1] Present address: COMET-NIRIM, National Institute for Research in Inorganic Materials, Tokyo Institute of Technology, c/o Professor H Koinuma, 4259 Nagatsuda, Midori-ku, Yokohama, Kanagawa 226-8503, Japan.

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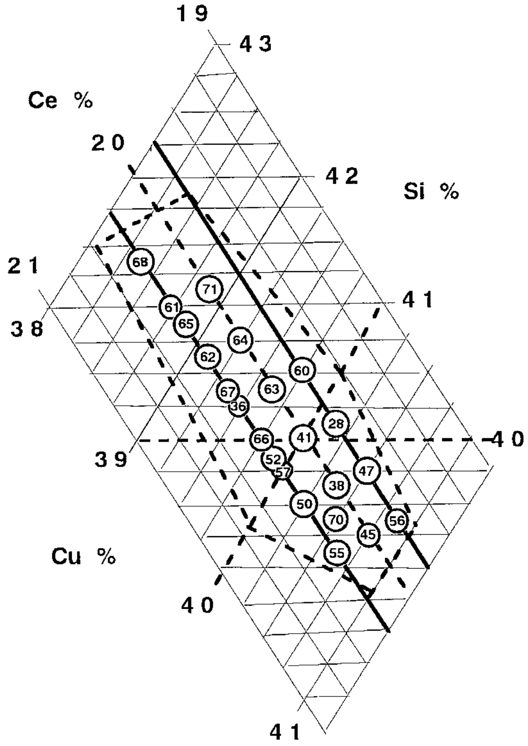
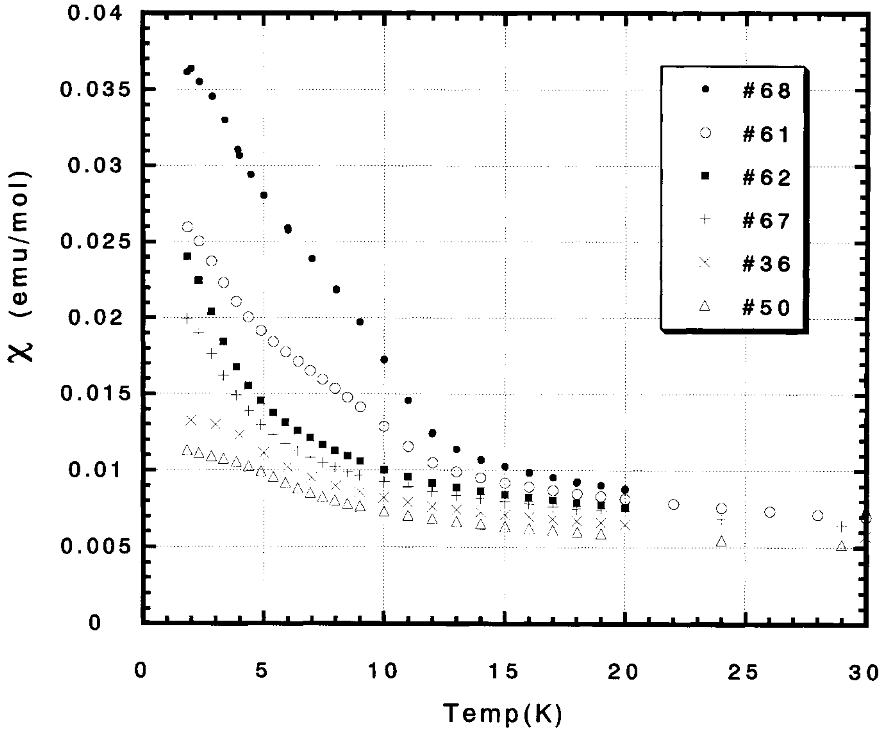


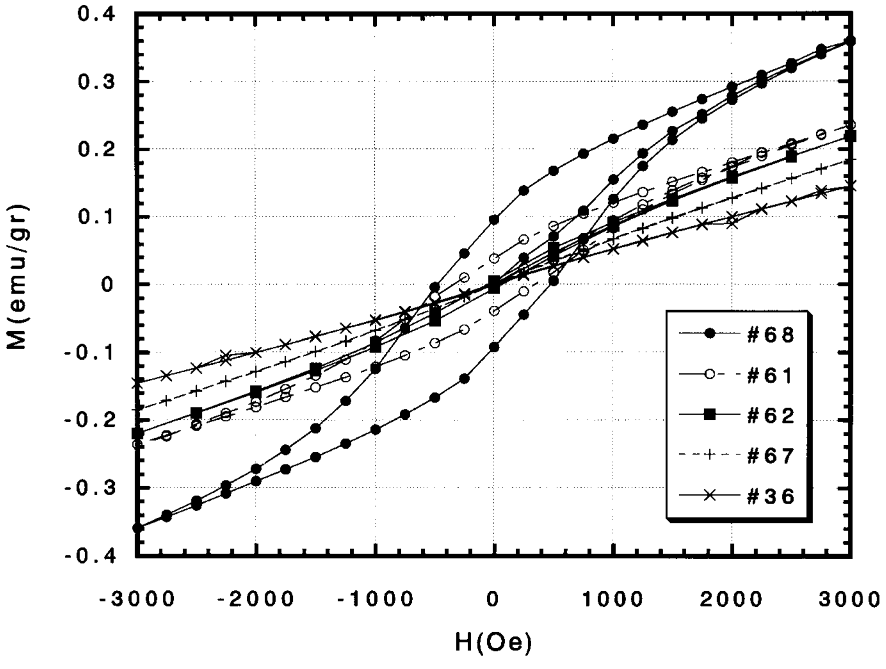
Figure 1. The homogeneity range of CeCu$_2$Si$_2$ determined at 750 °C [2] and the numbered samples prepared for this study.

two ferromagnetic samples (No 71 and No 64) were studied. Therefore, there seems to be a boundary of ferromagnetism and paramagnetism somewhere in between Cu 39.25 and 39.5 at.% and the superconducting phase emerges at the boundary. In addition to this systematic change of magnetic nature with composition, the existence of the peak around $2\theta = 33°$ does not necessarily correspond to the appearance of ferromagnetic signals; also the reannealing at 750 °C extinguished the impurity diffraction peak but not the ferromagnetism. From these facts we conclude that the ferromagnetism is not due to the impurity phase corresponding to the single weak line detected by means of x-ray diffraction nor to any other parasitic phases. Figure 3(a) displays the variation of the magnetic behaviour reflected in the specific heat. It is noted here that there is not a noticeable peak at about 12 K where the weak ferromagnetic signal appears for the susceptibility and magnetization, although a small anomalous background seems to appear below this temperature. A distinct broad peak develops only below 4 K and the peak broadens and diminishes with the increasing Cu content, shifting to lower temperatures. This last fact is very important for guaranteeing that the ferromagnetism is due not to a parasitic ferromagnetic phase, but to an intrinsic one of the compound, presumably due to heavy quasiparticles of the Kondo compound, although the precise nature is not known at present. This must be caused as a result of the change of the carrier density due to the Cu substitution for Si in the present case.

In figure 3(b), we now show the superconducting transition in the C_p/T versus T plot ($H = 0$) for several Cu-rich samples. We note that the discontinuity at T_c is very large ($[\Delta C/\gamma T]_{T_c} \approx 1.34$ for No 50 and No 70) and sharp for a polycrystalline sample, and that the others reveal a smaller and broader peak at T_c, although the latter group has a higher T_c ($\sim$0.7 K)

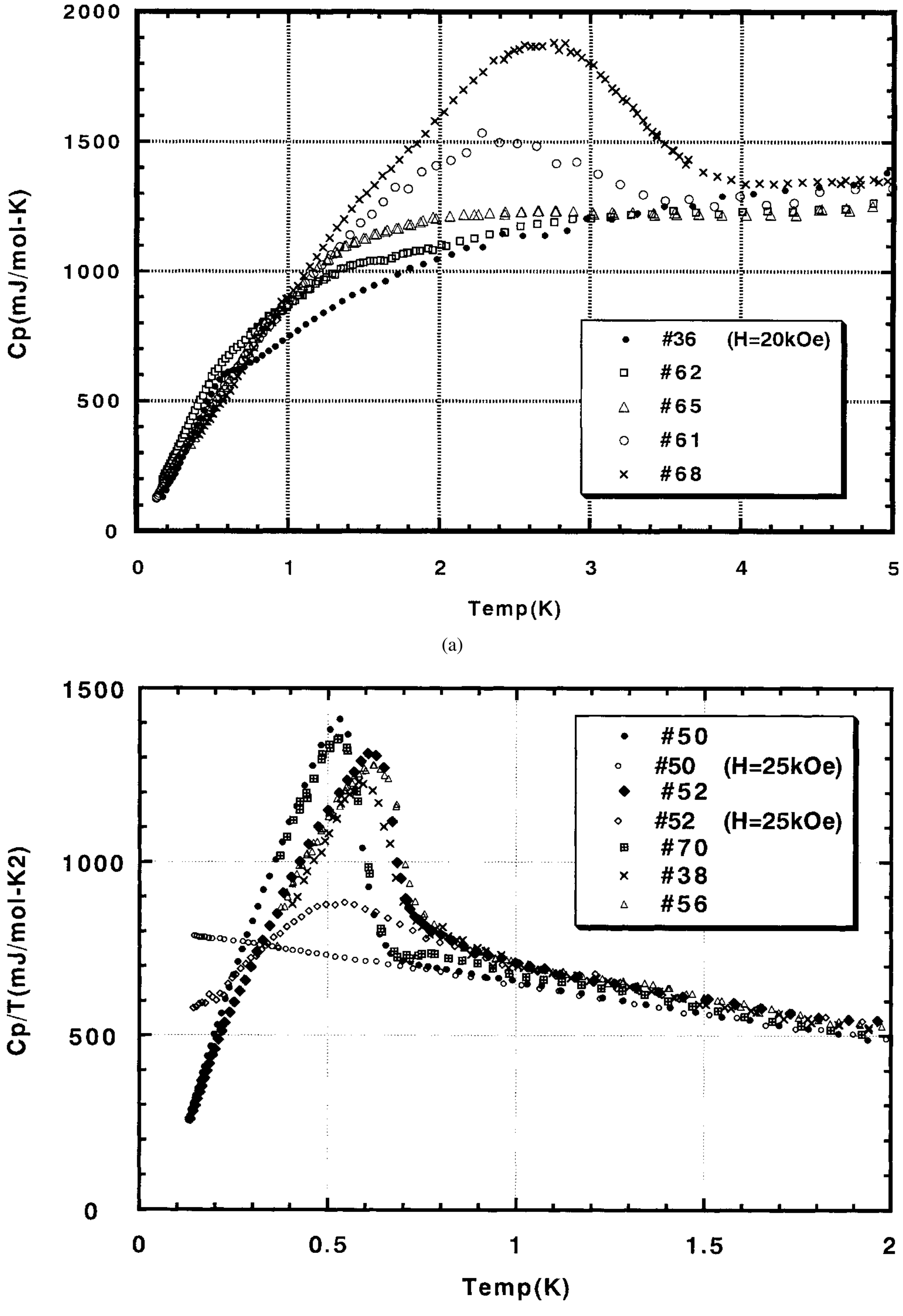


(a)



(b)

Figure 2. (a) Magnetic susceptibility and (b) M–H curves of several samples along the Ce 20.25% line, showing ferromagnetic behaviour below about 12.5 K.



(a)

(b)

Figure 3. (a) Specific heat curves showing the change of the magnetic peak below 4 K along the Ce 20.25% line and (b) C_p/T versus T showing two different superconducting transition temperatures (~0.6 and ~0.7 K).

than the former (~0.6 K). In any case, this seems to be a somewhat puzzling but genuine fact revealed in the present study. We will also show below two different types of behaviour in the resistivity corresponding to these two types of specific heat behaviour. We further demonstrate different specific heat behaviours of these two groups in figure 3(b), where the normal-state specific heat of No 50 and No 52 revealed under a magnetic field of 25 kOe is also displayed as a typical example for each group. The latter sample shows a broad peak around 0.5 K, while the former shows only an almost T-linear C/T down to 0.15 K, which was the limit of the present measurement. The broad peak around 0.5 K in the normal-state specific heat has been known for a while and is believed to be a signature of the somewhat mysterious phase 'A' [4]. It may, however, be instead a residue, probably of short-ranged type, of the magnetic order discussed above and the almost T-linear behaviour must be intrinsic to the superconducting phase, as reported for some single crystals [5]. It is remarked that the almost T-linear part of our data was successfully fitted up to about 1 K with the equation $C = \gamma T + \Gamma T^3 \ln T$, which has been known to apply for the specific heat due to spin fluctuations in, for example, the normal liquid ^{3}He [6].

We now present the results of resistivity measurements carried out on several typical samples. We reconfirmed the usual thermal variation with broad peaks around 100 K and 10 K, and a rapid decrease below the lower peak [7]. It is, however, worthwhile to comment on a new correlation between the residual resistivity just above T_c and the two values of T_c discussed above. We generally found that the higher-T_c group has a much smaller residual resistivity than the lower-T_c one: about 20 $\mu\Omega$ cm and 100 $\mu\Omega$ cm, respectively. This correlation seems to be a bit surprising, because the specific heat anomaly at T_c implies that the superconducting state of the lower-T_c group is more sturdy, with a larger volume fraction of the superconducting part, than that of the other, as already commented on above with respect to figure 3(b). A similar correlation seems to hold also for the resistivity data for single crystals [8]. We do not at present have any good interpretation for this correlation, but it may become an important clue to our understanding of the superconductivity of $CeCu_2Si_2$, for example, in relation to atomic disorder, as recently suggested by Louca *et al* [9]. It is important to note that these two groups of superconducting samples also reveal quite contrasting resistive behaviours in external magnetic fields, as displayed in figure 4. The upper case shows usual resistive behaviour of an ordinary superconductor in fields for a high-T_c sample, while the resistive variation shown in the lower case for the low-T_c sample (No 50) is unusual, because the resistivity in fields, say 22 and 23 kOe, increases again at lower temperatures, indicating that the superconductivity in such fields for some reason becomes unstable. Such anomalous T-dependence has been reported for magnetic superconductors before—for which, however, long-range magnetic order was responsible [10]. In the case of No 50, no phase transition such as magnetic ordering has been detected down to 0.06 K in the specific heat measurement, at least in zero magnetic field, performed in a nuclear demagnetization cryostat. We, therefore, emphasize that this resistive behaviour in the intermediate fields of the clean sample No 50 must be intrinsic to the superconductivity of the compound, as such behaviour was reported for a single-crystalline sample before [8].

In summary, we have carefully reinvestigated the heavy-fermion superconductor $CeCu_2Si_2$ by employing very homogeneous and well characterized polycrystalline samples and found the following new results:

(1) There exists a weakly ferromagnetic phase in the Cu-deficient region in the homogeneity range.
(2) The superconducting phase emerges upon disappearance of the ferromagnetic phase.
(3) There seem to be two distinct groups of superconducting phases, one with a lower T_c of

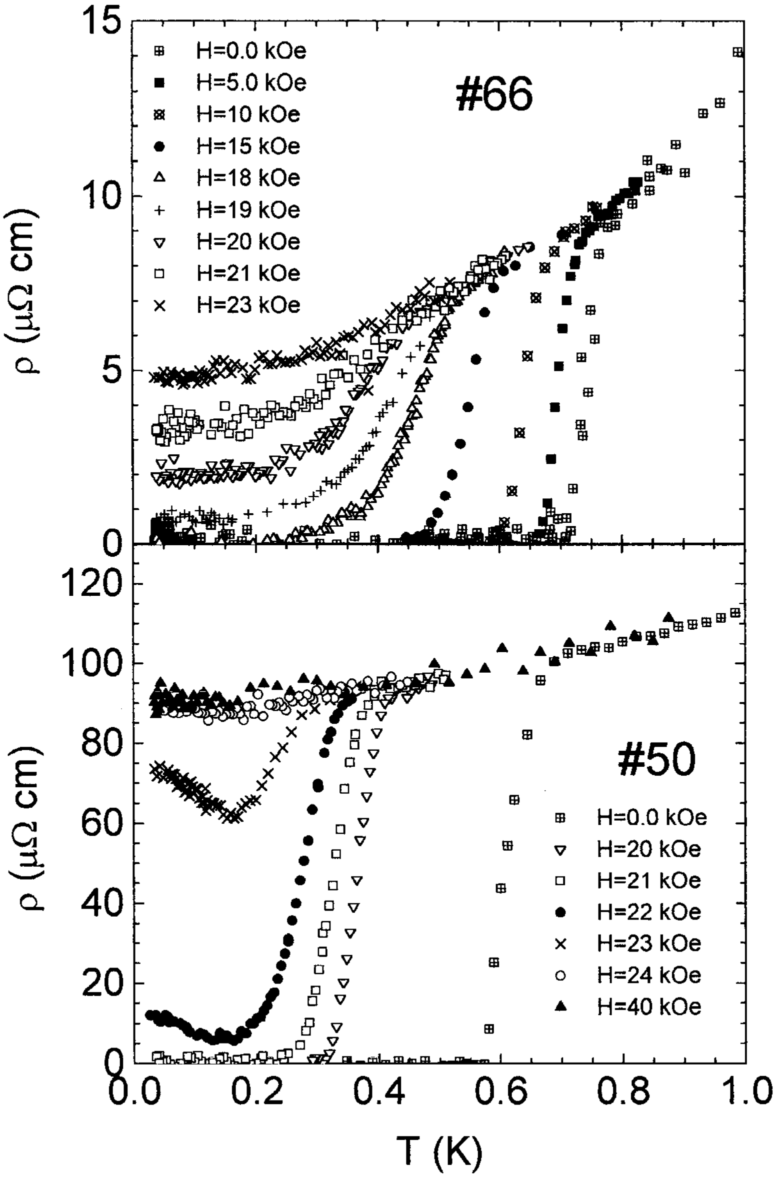


Figure 4. Two different resistivity behaviours in external magnetic fields, for samples No 66 (upper case) and No 50 (lower case) (see the text).

about 0.6 K and a high residual resistivity value of about 100 $\mu\Omega$ cm, the other with a higher T_c of about 0.7 K and a lower residual resistivity value of about 20 $\mu\Omega$ cm.

(4) The lower-T_c group of samples revealed anomalous resistive behaviour in intermediate magnetic fields and increasing resistivity with decreasing temperature.

The present system would be thereby the first case in which superconductivity emerges in such ferromagnetic surroundings under normal pressure, providing us with a unique opportunity to study the detailed interplay between superconductivity and ferromagnetism.

In view of this, it is noteworthy that an itinerant ferromagnet, UGe_2 has recently been reported to undergo a superconducting transition under pressure of about 1.3 GPa [11].

It is very important to investigate the heavy-fermion superconductor $CeCu_2Si_2$ further by employing well characterized samples in order to clarify the real nature of the compound—including the role of ferromagnetic interaction, the symmetry of pairing of the superconducting state and so forth. A more detailed account of the present work will be published soon elsewhere.

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