Effect of impurity scattering on the superconductivity of CeCu$_2$Si$_2$

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Abstract. The archetypal heavy fermion superconductor CeCu$_2$Si$_2$ exhibits an unusual pressure–temperature ($P$–$T$) phase diagram, in which superconductivity survives over a broad region in pressure (>10 GPa) and the superconducting transition temperature $T_c$ follows a complicated pressure dependence. To understand these unique properties, in this paper, we study a series of Ge-substituted single crystals CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$ ($x = 0.01, 0.1$ and $0.25$). It is found that superconductivity is significantly weakened due to the elevated impurity scattering resulting from the partial Ge/Si substitution. For the sample with $x = 0.01$, a minimum of $T_c(p)$ is revealed around 3 GPa. Upon further increasing $x$, the continuous superconducting region in the pure compounds breaks up into two disconnected superconducting domes. Superconductivity vanishes at $x \simeq 0.25$. These findings suggest that two different superconducting states, one magnetic and the other charge density fluctuations mediated, merge in CeCu$_2$Si$_2$.

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1. Introduction

Still the most significant example of electronic order in metals is the phenomenon of superconductivity. Many complex materials, including cuprate, organic and heavy fermion superconductors, follow remarkably similar phase diagrams [1], in which superconductivity is closely linked to magnetism. Due to the strong local Coulomb repulsion present in these systems, the charge carriers are subject to a large mass enhancement (up to a factor of 1000), which accordingly reduces the Fermi velocity \( v_F \). Therefore, the formation of superconductivity goes beyond the framework of the conventional BCS model since the retardation needed between the dynamics of the charge carriers and the ions of the crystal lattice (characterized by sound velocity \( v_s \)), \( v_F \gg v_s \), is not fulfilled. The true nature of this kind of superconductivity remains enigmatic.

In the last decade, a growing number of heavy fermion compounds, especially the Ce-based systems, have been found to follow a qualitatively similar pressure–temperature (\( P-T \)) phase diagram [2]–[4]: at or close to a quantum critical point (QCP), where the antiferromagnetic (AFM) ordering temperature \( T_N \) is continuously suppressed by applying pressure, superconductivity increases and the behaviour of the normal state deviates from Landau–Fermi liquid theory, which can nicely describe most simple metals and alloys at low temperature. These phenomena suggest that spin fluctuations contribute to the glue attracting electrons to form copper pairs in heavy fermion systems [3]. However, the archetypal heavy fermion superconductor CeCu_2Si_2 [5] and its isoelectronic compound CeCu_2Ge_2 revealed much more complicated behaviour under pressure [6, 7]. While an analogous magnetic QCP has by now been demonstrated in CeCu_2Si_2 at ambient pressure [8], the associated superconducting region extends to much higher pressure than in other compounds, reaching up to 10 GPa in some cases. In particular, \( T_c \) shows an unusual pressure dependence: \( T_c (\approx 0.7 \text{ K}) \) is nearly constant below 2 GPa, followed by a steep increase at 2–3 GPa and reaching a maximum value of about 2.2 K around 3 GPa, far away from the magnetic QCP [6]. These properties appear to contradict the magnetic interaction model.

To understand the unconventional nature of superconductivity in CeCu_2Si_2, we have prepared a series of Ge-substituted single crystals CeCu_2(Si_1-xGe_x)2. The partial substitution of Si by Ge in CeCu_2Si_2 has two effects: disorder and lattice expansion. The increased disorder scattering shortens the mean free path \( \ell \) and critically affects the occurrence of unconventional superconductivity, whereas the widening of the lattice may weaken the coupling between the conduction electrons and the localized 4f electrons of Ce and, therefore, favours long-range magnetic ordering at low temperature, providing an opportunity to study the magnetic properties in greater detail than previously possible. Compensating this volume increase by applying...
hydrostatic pressure then allows us to study essentially the same material but with a higher level of impurity scattering and to investigate the effects this may have on its superconducting properties. Recently, we have reported the observation of two distinct superconducting phases in CeCu$_2$(Si$_{0.9}$Ge$_{0.1}$)$_2$ [9, 10]. In this paper, we present a detailed description on how the superconducting phase evolves with increasing Ge-content in CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$ by measuring the electrical resistivity $\rho(T)$ under pressure.

The paper is organized as follows. In section 2, the experimental methods are described. In section 3, we present the $P$–$T$ phase diagrams for the samples with $x = 0.01$, 0.1 and 0.25, and discuss how the enhanced disorder scattering suppresses the occurrence of superconductivity. Finally, section 4 summarizes the main findings.

2. Experimental methods

Single crystals of the CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$ series were grown with excess Cu as flux medium in an aluminium-oxide crucible (40 mol% Cu + 60 mol% CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$ polycrystals). The growth speed was typically about 1 mm h$^{-1}$. With this technique, large single crystals with masses up to 2 g have been obtained. The powder x-ray diffraction measurements demonstrated that CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$ crystallizes in the ThCr$_2$Si$_2$ structure (I4/mmm), in which the Ce atoms take up body-centred tetragonal positions.

The resistivity was determined by low-current ac four-point measurements in an adiabatic demagnetization cryostat (down to $T = 180$ mK) and a dilution refrigerator (down to $T = 50$ mK). To achieve high pressure, two different pressure techniques were employed. The first one is the piston–cylinder cell, in which a 1:1 mixture of iso-pentane and n-pentane is filled as a hydrostatic pressure medium. With this technique, we can obtain a maximum pressure of about 3.5 GPa and simultaneously measure multiple samples (up to seven) in one pressure cell. To reach higher pressure, we changed to the Bridgman-type anvil cell with a solid pressure medium (steatite). Except for one sample with $x = 0.1$ (#1e), which was measured in the Bridgman-type cell, all the other measurements were performed in the piston–cylinder cells. Absolute resistivity values for the very small single crystals measured were obtained by extrapolating the high-pressure, room temperature data to zero pressure to match the ambient-pressure, room temperature resistivity measured on large reference crystals. The pressure was determined at low temperatures from the superconducting transition temperature $T_c$ of Sn (piston–cylinder cell) [11] and Pb (Bridgman anvil cell) [12].

3. Results and discussion

The crystal with $x = 0.1$ is particularly important for understanding the unusual superconducting phase diagram of CeCu$_2$Si$_2$ [9, 10]. At ambient pressure, the measurements of thermodynamic properties revealed that CeCu$_2$(Si$_{0.9}$Ge$_{0.1}$)$_2$ undergoes a transition at $T_N \approx 1.3$ K and a following first-order transition at $T_1 \approx 0.85$ K. Recent neutron diffraction experiments indicate an AFM transition at $T_N$ and a possible reorientation of the magnetic moments at $T_1$ [13]. Bulk superconductivity occurs at $T_c \approx 0.12$ K. By applying hydrostatic pressure, the transitions at $T_N$ and $T_1$ are gradually suppressed. In contrast to pure CeCu$_2$Si$_2$ and CeCu$_2$Ge$_2$, two separate superconducting domes are observed in the $P$–$T$ phase diagram of CeCu$_2$(Si$_{0.9}$Ge$_{0.1}$)$_2$ (figure 1).
Figure 1. The $P$–$T$ phase diagram for CeCu$_2$(Si$_{0.9}$Ge$_{0.1}$)$_2$. The superconducting transition temperature $T_c$ is determined from the mid-point of the resistivity drop at the superconducting transition and the Néel temperatures ($T_N$ and $T_1$) are obtained from the derivative of resistivity with respect to temperature. Two superconducting regimes are observed under pressure. Magnetic order vanishes in the low-pressure superconducting regime. Inset: the pressure dependence of $T_c$ for three different samples cut from the same batch, showing little sample dependence. Different symbols represent different samples: (⊙) #1a; (∆) #1b; (⊗) #1c; (■) #1e.

The low-pressure superconducting region lies on the threshold of the AFM order, suggesting a magnetically mediated pairing. For superconductivity in the high-pressure region, a new pairing mechanism based on the density fluctuations has been proposed [9]. To make sure that these properties are intrinsic and sample-independent, we have investigated five samples with $x = 0.1$ (#1a–#1e) cut from the same batch. It confirms that all the studied samples display similar features under pressure. For comparison, in the inset of figure 1 we plot the pressure dependence of $T_c$ for three samples (#1a–#1c). The dashed line is an extrapolation of $T_c$ to zero temperature for sample #1b because in this pressure range no superconductivity is visible above the accessible temperature limit (~180 mK in the demagnetization cryostat). Qualitatively, all the samples exhibit a similar pressure dependence of $T_c$: $T_c$ increases with increasing pressure for $0 < p < 1$ GPa, reaching a maximum value of 0.3–0.4 K at $p \simeq 1$ GPa. For $1 < p < 3$ GPa, $T_c$ decreases monotonically with increasing pressure. It may be important to note that the two superconducting regimes in the $P$–$T$ phase diagram are determined by employing two different samples (#1a and #1e). This is only due to the experimental limitations as described in section 2. However, for both samples, no evidence of superconductivity is observed around 3 GPa down to the lowest temperature measured. This suggests that the physical properties and, in particular, the pressure dependence of $T_c$ of both samples are very similar.

The partial Ge/Si substitution would definitely introduce disorder into the materials, being the main source of impurity scattering. Nevertheless, secondary effects, such as increased Cu/Si site interchange and sample inhomogeneities, cannot be ruled out completely, which may broaden the superconducting transition and cause step-like superconducting transitions for some poor samples. Such a possible case is shown in figure 2(a) for sample #1b ($x = 0.1$). Generally,
the pressure dependence of superconductivity is consistent with other samples. The multi-step superconducting transition may be due to an inhomogeneous content of Cu in the sample. If one assumes that each step in the resistivity drop is caused by a certain Cu content, then one may even extract some useful information on how the impurity scattering (due to the exchange of Cu and Si sites) influences superconductivity, which then can be compared to the results determined in the samples with nominally different Ge concentrations. In figure 2(b), we plot $T_c$ versus pressure for sample #1b at different resistivity drops which are mainly chosen from the different steps of resistivity. From figure 2(b), one can see that the superconducting domain becomes narrower and that the maximum of $T_c(p)$ shifts to higher pressure when $T_c$ is taken at lower resistivity. This may indicate that, with increasing impurity scattering by means of modifying the local Cu or Ge content, $T_c$ is reduced and the existing region of superconductivity in the phase diagram diminishes. To further clarify this situation, in the following, we study two more samples with different Ge concentrations, i.e., the weakly doped $x = 0.01$ and the overdoped $x = 0.25$.

In figure 3, the pressure dependences of $T_N$ and $T_c$ are shown for CeCu$_2$(Si$_{0.99}$Ge$_{0.01}$)$_2$. Different from the samples with $x = 0.1$, only one magnetic transition is observed in this sample.
Figure 3. The $P$–$T$ phase diagram for CeCu$_2$(Si$_{0.99}$Ge$_{0.01}$)$_2$. $T_c$ shows a maximum value of $T^\text{max}_c \simeq 0.38$ K at $p \simeq 1$ GPa and a minimum of $T^\text{min}_c \simeq 0.12$ K at $p \simeq 3$ GPa.

Figure 4. The electrical resistivity $\rho(T)$ of CeCu$_2$(Si$_{0.75}$Ge$_{0.25}$)$_2$ (sample #2a) at various pressures. $T_N$, as indicated by the arrows, is suppressed by applying pressure. No superconductivity is observed at any pressure ($T > 0.05$ K). Inset: the temperature derivative of the resistivity $d\rho/dT$, which sharply increases upon cooling to below $T_N$ for $p < 1$ GPa but decreases for $p > 1$ GPa.

Again, the Néel temperature $T_N$ is depressed under pressure. With regard to superconductivity, $T_c$ is still continuous over a broad pressure region for this weakly doped sample, but with a pronounced minimum ($T^\text{min}_c \simeq 0.12$ K) at $p \simeq 3$ GPa, roughly where a steep increase in $T_c$ is observed in pure CeCu$_2$Si$_2$. With further increasing pressure, another maximum in $T_c(p)$ is expected in this sample.

Figure 4 shows the electrical resistivity $\rho(T)$ of CeCu$_2$(Si$_{0.75}$Ge$_{0.25}$)$_2$ at various hydrostatic pressures. The derivatives of $\rho(T)$ against the temperature at $p = 0$ and $p = 1.52$ GPa are plotted.
in the inset. At ambient pressure, the resistivity decreases rapidly as the temperature drops below $T_N \approx 1.9$ K. Upon increasing pressure, $T_N$ is shifted to lower temperature and the resistivity anomaly at $T_N$ becomes less pronounced. Around $p = 1$ GPa where $T_N \approx 1.1$ K, $\rho(T)$ starts to increase below $T_N$ compared to the extrapolation of $\rho(T)$ from higher temperature. This behaviour is very similar to what is observed in A-phase CeCu$_2$Si$_2$ [8]. Further increase in pressure completely suppresses the long-range magnetic order. The pressure-induced change of resistivity behaviour below $T_N$ could be attributed to the modification of the magnetic structure under pressure (e.g., change of the propagation vector and/or of the size of the ordered moment). For the samples with a larger unit cell volume, a long-range AFM order of localized moments may develop below $T_N$. The freezing out of spin-disorder scattering on localized magnetic 4f-spins may dominate the resistivity and result in a decrease of the resistivity below $T_N$. Upon compressing the lattice either by applying pressure or by reducing the Ge concentration, the AFM structure could change from a more localized type to an itinerant spin density wave (SDW) type, i.e., due to an increasing hybridization between Ce-4f and conduction electrons. In this case, the opening of a partial energy gap at the Fermi surface would reduce the number of charge carriers and result in a relative increase of the resistivity below $T_N$. Similar phenomena have been reported for both CeRu$_2$Si$_2$ [14] and A-type CeCu$_2$Si$_2$ [8], for which system the existence of a SDW has, in fact, been recently established by neutron diffraction experiments [15].

In figure 5, the Néel temperature $T_N$ is plotted as a function of pressure for CeCu$_2$(Si$_{0.75}$Ge$_{0.25}$)$_2$. The $T_N$ of both samples (#2a and #2b), which reveal two slightly different values at $p = 0$, decreases in parallel and monotonically over more than one decade in temperature upon increasing pressure. The extrapolation of $T_N$ (#2b) to zero gives a critical pressure of $p_{c1} = 2.3 \pm 0.2$ GPa. This provides direct evidence for the existence of an AFM QCP in the compound CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$. Presumably due to strong impurity scattering, no anisotropic superconductivity is observed even near the magnetic QCP down to 50 mK in CeCu$_2$(Si$_{0.75}$Ge$_{0.25}$)$_2$. 

**Figure 5.** The $P$–$T$ phase diagram of CeCu$_2$(Si$_{0.75}$Ge$_{0.25}$)$_2$. $T_N$ is continuously suppressed by pressure in both samples #2a and #2b. No superconductivity is observed even near the magnetic QCP down to 50 mK (see inset).
Based on the above experimental results, in figure 6 we propose a schematic phase diagram for CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$ to summarize the evolution of the $P$–$T$ phase diagram with increasing the Ge concentration. As shown in [9], the pressure shift on the abscissa by $p_{c1}(x)$—the critical pressure at the AFM QCPs whose value increases with the Ge concentration—causes the values of $T_N$ from different materials to coincide, suggesting that, as far as magnetism is concerned, the replacement of Si by Ge can be offset by hydrostatic pressure. This allows us to regard the abscissa as a density (or volume) scale. The occurrence of superconductivity, however, is not only determined by volume. $T_c$ appears to be very sensitive to disorder introduced by partial Ge/Si substitution. For the pure compounds CeCu$_2$Si$_2$ and CeCu$_2$Ge$_2$ [6, 7], superconductivity exists continuously over a broad pressure range, showing a nearly pressure-independent $T_c$ below 2 GPa which is then followed by a sharp increase. As a small amount of Si is replaced by Ge ($0 < x < 0.1$), $T_c$ is reduced and its pressure dependence shows a valley in between two peaks. Upon further increasing $x$, the continuous superconducting region breaks up into two disconnected superconducting domes. In comparison with the pure compounds, the low-pressure superconducting dome corresponds to the region where $T_c$ is nearly constant and the high-pressure superconducting dome is associated with the region where $T_c$ goes through a maximum. Finally, superconductivity disappears in the highly substituted materials (e.g. $x = 0.25$). These properties indicate the existence of two distinct superconducting states in CeCu$_2$Si$_2$. Superconductivity in the low-pressure regime occurs around a magnetic QCP at $p_{c1}$, where $T_N \rightarrow 0$ and non-Fermi-liquid behaviour is observed in the normal state, compatible with theoretical predictions of the magnetic-interaction model. Far away from the magnetic QCP, a weak volume collapse was observed in stoichiometric CeCu$_2$Ge$_2$ at $p_{c2}$ ($\approx 15$ GPa) where $T_c$ reaches a maximum value [16]. The occurrence of superconductivity in the high-pressure regime might be related to this weak first-order symmetry-conserving volume-collapse transition, suggesting a novel pairing mechanism based on charge density fluctuations [9, 17, 18].

For unconventional superconductivity with an anisotropic order parameter, the quasiparticle mean free path $\ell$ must exceed the superconducting coherence length $\xi$ [3, 19]. For the system CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$, the coherence length $\xi$ shows a minimum value around the QCP and increases on either side. As an example, in figure 7, we present the pressure dependence of $\xi$ and $\ell$ for

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**Figure 6.** A schematic phase diagram for CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$. 
Figure 7. The pressure dependence of the mean free path $\ell$ and the coherence length $\xi$ for CeCu$_2$(Si$_{0.9}$Ge$_{0.1}$)$_2$ (sample #1d). Inset: the ratio of $\ell/\xi$ versus $T_c$. Around the magnetic QCP ($p_{c1} \simeq 1.5$ GPa), $\ell$ displays a maximum and $\xi$ a minimum. $T_c$ monotonically increases with increase in $\ell/\xi$.

the sample with $x = 0.1$ (#1d), where the coherence length $\xi$ and the mean free path $\ell$ are estimated from the initial slopes of upper critical field $B_{c2}$, $T_c$ and the residual resistivity $\rho_0$, respectively [20]. As an approximation, we assume a constant Fermi wave vector of $k_F = 1$ Å$^{-1}$ for all the samples under pressure, as was determined for pure CeCu$_2$Si$_2$ at ambient pressure in the clean limit [21]. A minimum of $\xi(p)$ and a maximum of $\ell(p)$ are observed around the magnetic QCP at $p_{c1} \simeq 1.5$ GPa, and $T_c$ increases monotonically with increase of the ratio of $\ell/\xi$ (see the inset). In figure 7, the mean free path $\ell$ derived from $\rho_0$ is generally shorter than $\xi$. It is noted here that one cannot seriously rely on the absolute values of $\ell$ due to the following factors: (1) The scaled resistivity $\rho(T)$ may be inaccurate. In CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$, the resistivity behaviour at high temperature strongly depends on the sample orientation [20]. In our experiments, we adopted a spot-welding technique to make electrical contacts between the thin leads and the tiny samples, for which it is usually difficult to control the sample orientation accurately. Therefore, the scaling would bring errors to the resistivity (a maximum factor of about 2). Moreover, the simple assumption of $k_F = 1$ Å$^{-1}$ is quite rough. Both factors may affect the accuracy of the mean free path $\ell$. (2) Sample inhomogeneity: sections of the sample with a long enough mean free path $\ell$ can become superconducting. For inhomogeneous samples, one may find small superconducting portions even if $\ell < \xi$ in most of the sample. This may explain the partial transitions observed in some samples we studied. (3) The mean free path $\ell$ of the heavy quasiparticles could be longer than that of the light quasiparticles. In our calculation, the obtained $\ell$ should reflect the charge transport, which is dominated by the light quasiparticles. However, the heavy quasiparticles form the Cooper pairs in the unconventional superconductivity. To compare with $\xi$, we need the mean free path $\ell$ coming from the scattering of heavy quasiparticles. It was found that, in pure CeCu$_2$Si$_2$, the mean free path for spin–orbit scattering ($\ell_{so}$ $\simeq$ 200 Å, derived from the fit of $B_{c2}$) is clearly larger than the total mean free path ($\ell \simeq 20$ Å) [21]. All these suggest that the relation of $\ell/\xi > 1$ would be plausible for the superconducting sections. The purpose of plotting $\ell(p)$ in figure 7 is to show the trend of the mean free path $\ell$ under pressure, which should be reliable here. Around the QCP, both $\xi$ and $\ell$ favour the occurrence of superconductivity. For the Ge-doped materials...
Figure 8. A model for discussing the influence of impurity scattering on superconductivity. For unconventional superconductivity, the mean free path $\ell$ must exceed the coherence length $\xi$ [3, 19]. In the pure compounds, $\ell$ is large so that superconductivity can survive over a broad pressure range. The partial Ge/Si substitution leads to the reduction of $\ell$, restricting the occurrence of superconductivity to a narrow pressure range near the quantum critical point.

where $\xi \sim \ell$, superconductivity may be cut off as the system is tuned away from the magnetic QCP. Similar arguments should be applicable to explain the second dome of superconductivity in the $P-T$ phase diagram at high pressure. Since in this regime, superconductivity is strongly suppressed with increasing impurity scattering (i.e., the Ge concentration $x$) and the normal state reveals non-Fermi-liquid behaviour, superconductivity may be of an unconventional nature as well. To elaborate the physical properties in the high-pressure regime, further experiments are in progress.

Concerning the impurity scattering, $\ell$ is larger in the pure compounds CeCu$_2$Si$_2$ and CeCu$_2$Ge$_2$ than in the partially Ge-substituted materials CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$. From the schematic drawing shown in figure 8, one expects that superconductivity would survive over a broader pressure range in the pure compounds than in the doped cases. These arguments suggest that two distinct superconducting phases may intrinsically exist in the $P-T$ phase diagram of CeCu$_2$Si$_2$. However, the two relatively broad superconducting domes overlap with each other and therefore result in one continuous and broad superconducting region. The effect of pair breaking due to the increased impurity scattering, e.g., by partial Ge/Si substitution, then allows the observation of two separate superconducting regimes.

4. Conclusion

We have studied the superconducting phase diagrams for the Ge-substitution series CeCu$_2$(Si$_{1-x}$Ge$_x$)$_2$ ($x = 0.01, 0.1$ and $0.25$) by measuring the resistivity $\rho(T)$ under pressure. It is found that superconductivity is significantly weakened by partially substituting Si with Ge. The unusually broad superconducting region in the $P-T$ phase diagram of pure compounds CeCu$_2$Si$_2$ and CeCu$_2$Ge$_2$ breaks up into two separate superconducting domes with increasing...
Ge-content x. For the samples with x = 0.25, no superconductivity is observed even close to the AFM QCP. The effect of impurity scattering on unconventional superconductivity is discussed. The prerequisite for the occurrence of superconductivity with anisotropic gap function is that the mean-free path \(\ell\) must exceed the coherence length \(\xi\). In CeCu\(_2\)(Si\(_1-x\)Ge\(_x\))\(_2\), the upper critical field measurements reveal that \(\xi\) has a minimum near the AFM QCP and increases on either side, indicating that superconductivity may be cut off when the system is tuned away from the QCP. These findings suggest that two superconducting states, spin-fluctuation-mediated superconductivity in the low-pressure region and an enigmatic—possibly charge-density fluctuation mediated—superconducting state in the high-pressure region, merge in stoichiometric CeCu\(_2\)Si\(_2\).

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