Do antiferromagnetism and superconductivity coexist in 2% and 10% Ge doped CeCu$_2$Si$_2$?

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Abstract. Doping the heavy-fermion compound CeCu$_2$Si$_2$ with Ge allows for studying the unconventional superconducting state in the presence of a more stabilized antiferromagnetic phase. We performed heat capacity and elastic neutron scattering measurements on single crystals of 2% and 10% Ge doped CeCu$_2$Si$_2$ at different temperatures and magnetic fields. The zero field superconducting and magnetic transition temperatures in 2% Ge doped CeCu$_2$Si$_2$ are still close to those in pure CeCu$_2$Si$_2$, whereas $T_c$ and $T_N$ differ by an order of magnitude in 10% Ge doped CeCu$_2$Si$_2$. Our neutron scattering results show that the magnetic Bragg peak intensity decreases with decreasing temperature below $T_c$ in CeCu$_2$(Si$_{0.98}$Ge$_{0.02}$)$_2$. However, in CeCu$_2$(Si$_{0.9}$Ge$_{0.1}$)$_2$, it stays constant in the superconducting regime down to lowest temperatures. We conclude that, while a phase separation between magnetic and superconducting volumes takes place in 2% Ge doped CeCu$_2$Si$_2$, antiferromagnetism and superconductivity coexist on the microscopic scale in 10% doped CeCu$_2$Si$_2$.

1. Introduction

The interplay of superconductivity and magnetism in the heavy-fermion compounds is of fundamental interest in condensed matter physics. In CeCu$_2$Si$_2$, the first discovered heavy-fermion superconductor [1], this interplay is especially complex, and despite extensive research over the past years, many open questions still persist.

The ground state properties of CeCu$_2$Si$_2$, which crystallizes in the tetragonal ThCr$_2$Si$_2$ structure, depend very sensitively on the exact stoichiometry within the homogeneity range and on the growing conditions. While a slight Cu excess yields superconducting samples (S-type), Si rich ones show antiferromagnetic order (A-type). Samples with a composition very close to the nominal stoichiometry exhibit both properties (A/S-type) [2]. After it became possible to grow large single crystals with well-defined properties, neutron scattering studies in particular have contributed new insights. Thus the nature of the A-phase was found to be antiferromagnetic order of incommensurate spin-density-wave type [3]. In A-type CeCu$_2$Si$_2$ the ordered magnetic moment is $\mu_{\text{ord}} \approx 0.1 \mu_B$ at temperatures well below $T_N = 800$ mK. The propagation vector of the spin-density wave $\tau = (0.215 0.215 0.53)$ is determined by the nesting properties of the Fermi surface of the heavy quasiparticles, as calculated with the renormalized band method [3].
Below $T \approx 350$ mK the propagation vector is locked-in, while it shows a weak temperature dependence above.

A/S-type CeCu$_2$Si$_2$ exhibits a transition into the antiferromagnetically ordered phase at $T_N \approx 700$ mK, the propagation vector being approximately the same as in pure A-type samples, and a superconducting transition at $T_c \approx 500$ mK. Neutron scattering and $\mu$SR measurements provide evidence that the superconducting volume fraction grows at the expense of the magnetic one upon decreasing the temperature [4, 5]. This means that superconducting regions supersede magnetically ordered regions, and there is no microscopic coexistence of both phenomena.

Neutron scattering experiments on S-type CeCu$_2$Si$_2$, where $T_c \approx 600$ mK, revealed elastic short-range magnetic correlations with correlation lengths of approximately 50-60 Å in the superconducting state [6]. These appear at the positions of the magnetic Bragg peaks in the magnetically ordered representatives ($\mathbf{Q}_{AF}$). Furthermore, while the low-energy response is quasielastic above $T_c$, an inelastic response was observed at $\mathbf{Q}_{AF}$ in the superconducting state [6]. The size of the spin-excitation gap is $\Delta \approx 0.22$ meV. This is comparable to $4 k_BT_c$, a value close to that expected for the superconducting energy gap in BCS theory.

Ge atoms are isoelectronic with Si, but have a larger volume. Therefore, upon Ge doping, the hybridization between Ce atoms is decreased, which results in successive suppression of superconductivity and enhancement of the magnetic order. Hence, modest Ge doping allows for studying superconductivity in the presence of a more stabilized magnetic phase. Cu-NQR and $\mu$SR measurements on polycrystalline samples of 2% and 10% Ge doped CeCu$_2$Si$_2$ gave indications of coexistence of both properties [7]. The aim of our investigations was to check these results by means of elastic neutron scattering on single crystals.

2. Experiment and results

The experiments were conducted on high-quality single crystals of CeCu$_2$(Si$_{0.98}$Ge$_{0.02}$)$_2$ ($m = 0.75$ g) and CeCu$_2$(Si$_{0.9}$Ge$_{0.1}$)$_2$ ($m = 1.9$ g), both grown out of copper flux. On CeCu$_2$(Si$_{0.98}$Ge$_{0.02}$)$_2$ elastic neutron scattering measurements were performed on the three-axis spectrometer IN12 at Institut Laue-Langevin, Grenoble. The neutron wave vector was fixed to $k = 1.15$ Å$^{-1}$. The sample was mounted with the [1 1 0] direction vertical, which results in the [1 1 0]-[0 0 1] plane being the scattering plane. In order to track the onset of superconductivity, the ac susceptibility was logged simultaneously during the neutron scattering experiment.

Elastic neutron scattering measurements on CeCu$_2$(Si$_{0.9}$Ge$_{0.1}$)$_2$ were carried out on the three-axis spectrometer PANDA at Forschungsneutronenquelle Heinz Maier-Leibnitz, Garching, with a fixed neutron wave vector of $k = 1.185$ Å$^{-1}$, and again the [1 1 0]-[0 0 1] plane being the scattering plane. Both single crystals were characterized by heat capacity measurements applying the quasi-adiabatic heat-pulse method described in [8], which was slightly modified for the use of large samples.

Figure 1 displays the temperature dependence of the heat capacity divided by temperature at zero magnetic field between $T = 50$ mK and 2 K. There are two phase transitions in 2% Ge doped CeCu$_2$Si$_2$: Magnetic order sets in at $T_N = 750$ mK, and the sample becomes superconducting at $T_c = 500$ mK, which was verified additionally by measuring the ac susceptibility during the neutron scattering experiment. 10% Ge doped CeCu$_2$Si$_2$ orders magnetically below $T_N = 1.3$ K and becomes superconducting at $T_c = 130$ mK. Furthermore, there is a first order phase transition at $T_1 = 800$ mK. It could be ascribed to a lock-in transition of the propagation vector of the magnetic order. This lock-in transition is also present in CeCu$_2$(Si$_{0.98}$Ge$_{0.02}$)$_2$, but not pronounced enough to result in a distinct feature in the heat capacity.

In order to study the evolution of the magnetic order with temperature in the case of 2% Ge doped CeCu$_2$Si$_2$, we performed rocking scans by rotating the sample through $\mathbf{Q} = (0.208 0.208 1.482)$ ($\omega$-scan). In the case of 10% Ge doped CeCu$_2$Si$_2$ the exact $\mathbf{Q}$ position of the magnetic Bragg reflection was first determined by scanning the $h$ and $l$ component of the
expected $Q$ vector separately at each temperature. Afterwards a combined $\omega - 2\theta$ scan around the determined $Q$ was performed, where $2\theta$ is the scattering angle. Below $T_1$ the position of the magnetic Bragg peak is $Q = (0.227 \ 0.227 \ 1.492)$. Above $T_1$ the propagation vector is temperature dependent and takes a value of $Q = (0.238 \ 0.238 \ 1.458)$ at $T = 1.3$ K. The thus obtained data (intensity versus $\omega$) were fitted with peaks of Gaussian lineshape, the area of which is the integrated intensity. Figures 2 and 3 display this integrated magnetic intensity as a function of temperature for CeCu$_2$(Si$_{0.98}$Ge$_{0.02}$)$_2$ and for CeCu$_2$(Si$_{0.9}$Ge$_{0.1}$)$_2$, respectively.

In the case of 2% Ge doped CeCu$_2$Si$_2$ the integrated magnetic intensity increases with

![Figure 1](image1.png)

**Figure 1.** Temperature dependence of the heat capacity divided by temperature at $B = 0$ for CeCu$_2$(Si$_{0.98}$Ge$_{0.02}$)$_2$ (closed squares) and CeCu$_2$(Si$_{0.9}$Ge$_{0.1}$)$_2$ (open circles). Phase transition temperatures are marked by arrows.

![Figure 2](image2.png)

**Figure 2.** Temperature dependence of the integrated magnetic intensity in 2% Ge doped CeCu$_2$Si$_2$ at $B = 0$. The integrated intensity results from Gaussian line shape fits to rocking scans (intensity vs. sample rotation angle $\omega$) across the magnetic Bragg position.

![Figure 3](image3.png)

**Figure 3.** Temperature dependence of the integrated magnetic intensity in 10% Ge doped CeCu$_2$Si$_2$ at $B = 0$. The integrated intensity results from Gaussian line shape fits to measurements of the intensity vs. the sample rotation angle $\omega$ obtained from $\omega - 2\theta$ scans across the magnetic Bragg position.
decreasing temperature below $T_N$ and reaches its maximum around $T_c$. Below $T_c$ it decreases with decreasing temperature to a constant low value below $T \approx 300$ mK. In 10\% Ge doped CeCu$_2$Si$_2$ the integrated magnetic intensity increases with decreasing temperature below $T_N$ and does not decrease upon crossing $T_c$, but stays constant within the error bars in the superconducting regime.

3. Discussion

Our neutron scattering measurements on 2\% and 10\% Ge doped CeCu$_2$Si$_2$ show qualitatively different behaviour. The successive reduction of the magnetic intensity with decreasing temperature in the superconducting regime in CeCu$_2$(Si$_{0.98}$Ge$_{0.02}$)$_2$ indicates that the superconducting volume fraction grows at the expense of the magnetic one. This implies that spatial phase separation of antiferromagnetism and superconductivity takes place. Therefore, unlike Cu-NQR measurements on polycrystalline samples [7], our results do not support the scenario of microscopic coexistence of the two phenomena.

In CeCu$_2$(Si$_{0.9}$Ge$_{0.1}$)$_2$ the transition into the superconducting state is denoted by a small, but distinct peak in $C/T$. In comparison, a sharp superconducting transition was observed in the electrical resistivity on polycrystalline samples [9]. By measuring the Meissner effect, the superconducting volume fraction was determined to exceed 30\% [10], so superconductivity can safely be considered a bulk property. In contrast to the results on 2\% Ge doped CeCu$_2$Si$_2$, the observation that the magnetic volume fraction is not significantly diminished with decreasing temperature in the superconducting phase suggests that there is no separation of superconducting and antiferromagnetic volumes in 10\% Ge doped CeCu$_2$Si$_2$. It appears that superconductivity coexists with antiferromagnetism if the superconducting phase forms in a well ordered magnetic phase.

In conclusion, our elastic neutron scattering results on 2\% Ge doped CeCu$_2$Si$_2$ do not support the scenario of microscopic coexistence of superconductivity and antiferromagnetism, while elastic neutron scattering on 10\% Ge doped CeCu$_2$Si$_2$ points towards coexistence of the two phenomena on the microscopic scale. This might indicate that a more stabilized magnetism is favourable for its coexistence with superconductivity.

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4. References

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