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The phase diagram of $CeCu_2(Si_{1-x}Ge_x)_2$

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Abstract

The magnetic phase diagram of $CeCu_2(Si_{1-x}Ge_x)_2$ has been studied by means of neutron diffraction to explore the transition from heavy-fermion superconductivity to local moment antiferromagnetism. This system shows up to three distinct phase transitions below 5 K. The incommensurate magnetic structure of pure $CeCu_2Ge_2$ is amazingly stable against the application of a magnetic field. With increasing Si concentration, this structure is essentially preserved down to $x \approx 0.4$, where the magnetic moments are suppressed below the sensitivity of neutron powder diffraction. A further low-temperature phase transition is characterized by a spin reorientation. For $x \leqslant 0.4$, a different type of magnetism is established. The nature of this new type of magnetic ordering, which may also be present in $CeCu_2Si_2$, still has to be explored. A comparison is made with the magnetic phase diagram of $Ce(Cu_{1-x}Ni_x)_2Ge_2$.

1. Introduction

In $CeCu_2Ge_2$ the on-site Kondo-type and the inter-site RKKY-type interaction strength are of the same order of magnitude. Below $T_N = 4.15$ K the onset of long-range magnetic order is characterized by an incommensurate amplitude modulated structure of localized Kondo compensated magnetic moments of $\mu_{Ce} = 1.05 \pm 0.1\mu_B$ [1]. Single-crystal neutron diffraction showed that this magnetic structure is not altered by a magnetic field of $B = 8$ T applied along the a-axis at $T = 2.4$ K, thus exceeding the magnetic correlation energy $k_B T_N$ almost twice [2, 3]. Substituting Ge by Si increases the hybridization strength, as measured by the Kondo lattice temperature T^*, mainly by reducing the unit cell volume. As expected, this is accompanied by a reduction in the ordering temperature, as shown in the schematic phase diagram of $CeCu_2(Si_{1-x}Ge_x)_2$ in Fig. 1. For low Ge concentrations $x \leqslant 0.05$, no magnetic order, but a transition into a superconducting ground state has been detected. Resistivity and susceptibility measurements showed further phase transitions for intermediate concentrations around 2 (labelled T_2) and 0.5 K (labelled T_3) in Fig. 1 [4, 5].

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2. Experimental results and discussion

We have investigated the magnetic phase diagram by powder neutron diffraction for $x = 0.8, 0.6$ and 0.4 in the range $1.5\text{ K} \leqslant T \leqslant 300$ K. These experiments have been performed on the multidetector diffractometers E6 at the Hahn Meitner Institute, Berlin and D1B at the ILL, Grenoble. The magnetic structure of pure $CeCu_2Ge_2$ is essentially preserved for $x = 0.8$ and 0.6 between T_2 and T_N. The phase transition at T_2 is characterized by a spin reorientation, while the propagation vector of the incommensurate structure remains unchanged. The third transition around 0.5 K was out of range in these experiments. For $x = 0.4$, no

Table 1
Propagation vector and modulus of the magnetic moment at $T = 1.6$ K of $CeCu_2(Si_{1-x}Ge_x)_2$ as determined by neutron diffraction. For $x = 0.4$ and 0.2, no magnetic intensities have been observed

x	0.2	0.4	0.6	0.8	1
$\boldsymbol{q}$	—	—	(0.271, 0.271, 0.520)	(0.269, 0.269, 0.550)	(0.28, 0.28, 0.53)
Magnetic moment	—	—	$0.48 \pm 0.1\mu_B$	$0.59 \pm 0.1\mu_B$	$1.05 \pm 0.1\mu_B$

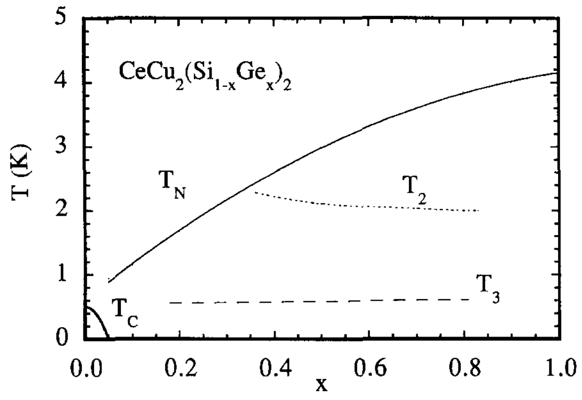

Fig. 1. Magnetic phase diagram of $CeCu_2(Si_{1-x}Ge_x)_2$. The solid line indicates the Néel temperature T_N. For intermediate concentrations two additional phase transitions can be seen at T_2 and T_3, respectively. T_c corresponds to the transition into the superconducting state.

magnetic intensities could be observed, leading to an estimation for the upper limit of the magnetic moments of $\mu \leqslant 0.25\mu_B$. The results are summarized in Table 1. We then tried to determine the magnetic structures of $x = 0.2$ and 0.4 by single-crystal neutron diffraction on the triple-axis spectrometer E7 at the Hahn Meitner Institute, Berlin. However, no magnetic intensities could be observed. This implies either a further reduction of the magnetic moments or a different magnetic structure for low Ge concentrations. Already, on the basis of macroscopic measurements [6], it has been concluded that the magnetic structures undergo significant modifications when passing through some critical concentration around $x = 0.4$. It is intriguing to compare $CeCu_2(Si_{1-x}Ge_x)_2$ with $Ce(Cu_{1-x}Ni_x)_2Ge_2$. Similar to the Ge–Si system, the substitution of Cu by Ni also compresses the unit cell and increases the hybridization [1]. However, already small Ni concentrations strongly suppress the magnetic ordering temperature [1]. This shows that in addition to the volume effect the change of the electronic structure plays an important role. Doping pure $CeCu_2Ge_2$ with a few at% of Ni results in a second magnetic phase transition. Based on the temperature dependence of the intensities of the principal magnetic reflections as determined by single-crystal neutron diffraction of $CeCu_{1.9}Ni_{0.1}Ge_2$, these two magnetic phases seem to superimpose independently of each other [7]. The corresponding two magnetic structures are very similar to each other as well as to pure $CeCu_2Ge_2$. Again, a magnetic field of $B = 8$ T applied along the a-axis leaves these two magnetic structures unchanged [3]. For higher Ni concentrations, a new type of magnetic order starts developing [1]. The now available single crystals enabled us to investigate this new magnetic phase in more detail. In contrast to the results obtained by powder neutron diffraction [1], single-crystal diffraction of $CeCuNiGe_2$ ($x = 0.5$) revealed a different magnetic structure characterized by a propagation vector $\boldsymbol{q} = (\frac{1}{2}, \frac{1}{2}, 0.81)$. For Ni concentrations above $x \geqslant 0.8$, long-range magnetic order is completely suppressed and deviations from Fermi-liquid behaviour have been detected [8].

To conclude, $CeCu_2(Si_{1-x}Ge_x)_2$ shows a complex magnetic phase diagram. The magnetic structures for $x = 0.8$ and 0.6 have been determined and are very similar to that of pure $CeCu_2Ge_2$. An additional low-temperature magnetic phase transition is characterized by a spin reorientation. For Ge concentrations $x \leqslant 0.4$, no magnetic intensities could be observed, neither by powder, nor by single-crystal neutron diffraction. The magnetic structure of pure $CeCu_2Ge_2$ (and 5% Ni doping) remains unchanged by the application of a magnetic field of $B = 8$ T. Further neutron scattering studies at very low temperatures and in dependence of an applied magnetic field are highly warranted.

Most importantly, the magnetic structures of the Si-rich compounds should be solved by single-crystal diffraction to contribute to the long-standing debate of a possible long-range magnetic order in the heavy-fermion superconductor $CeCu_2Si_2$.

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