Incipient superconductivity and NFL behavior in off-stoichiometric $Ce_{1+x}Ni_{2+y}Ge_{2+z}$ polycrystals

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Abstract

The influence of composition on the physical properties of $CeNi_2Ge_2$ was studied by measuring the resistivity of slightly off-stoichiometric $Ce_{1+x}Ni_{2+y}Ge_{2+z}$ polycrystals at low temperatures ($0.01\ K \leqslant T \leqslant 6\ K$). We establish a clear correlation between composition, residual resistivity ρ_0 and incipient superconductivity, i.e. a drop in $\rho(T)$. The lowest ρ_0 values and the largest reductions in resistivity, reaching $\rho = 0$ for one particular sample, were observed in slightly Ni-rich samples. A weak decrease of the unit-cell volume with increasing Ni-content suggests a slightly larger 4f-hybridization in those Ni-rich samples. The exponent ε of the $(\rho - \rho_0) \sim T^{\varepsilon}$ behavior found for $T \leqslant 2$ K correlates with the residual resistivity. While $\varepsilon = 1.5$ is observed for $\rho_0 \geqslant 3\ \mu\Omega$ cm, $1.3 \leqslant \varepsilon < 1.5$ was found for $0.17\ \mu\Omega\ \text{cm} \leqslant \rho_0 < 3\ \mu\Omega$ cm.

Keywords: Non-Fermi liquid behavior; Heavy-fermion superconductivity; Electrical resistivity; Quantum-critical point

Up to now $CeCu_2Si_2$ is the only Ce-based heavy-fermion system (HFS) that shows superconductivity (SC) already at ambient pressure. Its ground-state properties (SC and/or magnetic phase "A") depend sensitively on the strength of hybridization, g, between 4f- and conduction electrons [1]. By application of small pressure or a slight Cu-excess within the homogeneity range, the magnetic ordering temperature T_A could be suppressed to zero. This defines a quantum-critical point (QCP) g_c where pronounced non-Fermi liquid (NFL) effects are observed [2]. The non-magnetic homologue HFS $CeNi_2Ge_2$ shows very similar NFL behavior at $p = 0$ [3]. Therefore, a similar strong dependence of the ground-state properties on g can be expected. Since the increase of g by the application of hydrostatic pressure induces SC for $p \geqslant 1.7$ GPa [4] SC might also occur at ambient pressure if g could be increased by changes in composition. Indeed Grosche et al. [5] recently reported the observation of SC below 0.2 K at $p = 0$ in a small piece of a single crystal, whose exact composition, however, could not be determined. The aim of the present work is to systematically study the composition dependence of the ground-state behavior of $CeNi_2Ge_2$.

All slightly off-stoichiometric polycrystals whose starting compositions are plotted in the ternary phase diagram in Fig. 1, were prepared in the same way by melting the appropriate amounts of pure elements in an arc furnace. After annealing at 800°C for 120 h the lattice constants of the $CeNi_2Ge_2$ main phase were determined by X-ray diffraction. Subsequently, the resistivity of pieces of approximately $6 \times 2 \times 2\ mm^3$ was measured in a $^3He/^4He$ dilution refrigerator ($10\ mK \leqslant T \leqslant 6\ K$) using a four-terminal low-frequency (17 Hz) lock-in technique. Below 0.06 K the temperature was measured using a cerium-magnesium-nitrate thermometer that was calibrated against an NBS superconducting fixed-point device. Since the granular structure of our polycrystals prevents the determination of absolute values of ρ, we estimated ρ by multiplying $R(T)/R_{300\ K}$ with $\rho_{300\ K} \approx 60\ \mu\Omega$ cm [6].

We found a clear correlation between composition, lattice parameters, residual resistivity and the onset of SC. Small Ni-excess leads to a decrease of the c-axis

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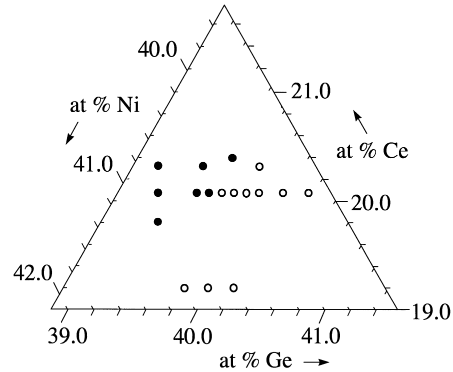

Fig. 1. Section of the ternary Ce–Ni–Ge phase diagram around 1-2-2 composition. Symbols mark starting composition for sample preparation. Due to losses of Ce during the preparation process the true Ce-content is expected to be about 2% lower. Closed circles mark samples that show an onset of SC below 0.1 K.

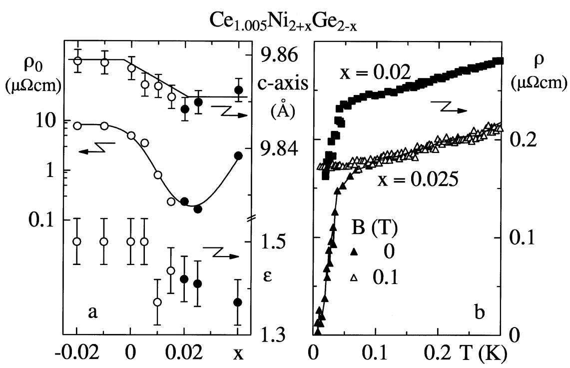

Fig. 2. (a) c-axis lattice parameter, residual resistivity ρ_0 as well as exponent ε in the observed $(\rho - \rho_0) \sim T^{\varepsilon}$ behavior for $T \leqslant 2$ K versus x for the series $Ce_{1.005}Ni_{2+x}Ge_{2-x}$. Closed circles represent crystals that show SC. (b) Low-T resistivity at $B = 0$ for polycrystals with $x = 0.02$ (■) and $x = 0.025$ (▲) showing incipient SC below 0.1 K Open symbols (△) show normal-state behavior for $x = 0.025$ at $B = 0.1$ T.

parameter, as shown in Fig. 2a for polycrystals of the series $Ce_{1.005}Ni_{2+x}Ge_{2-x}$. For the a-axis parameter no changes could be resolved. The resulting decrease of the unit-cell volume with increasing Ni-content should lead to a stronger 4f-hybridization in Ni-rich samples. The most interesting result of the present work is that the occurrence of incipient SC in the resistivity below 0.1 K is clearly related to the composition and occurs in the Ni-rich region of the phase diagram. Only for one of the samples studied so far with the composition $Ce_{1.005}Ni_{2.025}Ge_{1.975}$ $\rho = 0$ is reached (see Fig. 2b). The resistivity drop at T_c is suppressed either in magnetic fields $B \geqslant 0.1$ T or current densities $j \geqslant 0.05$ A/cm^2. This might suggest that we are dealing only with faint traces of SC of some Ni-rich foreign phase. However, the observation of the lowest residual resistivity of $\rho_0 = 0.17$ μΩ cm for this particular sample strongly supports an intrinsic nature of SC.

Finally, we discuss the apparent NFL behavior in $\rho(T)$, namely the power-law behavior $(\rho - \rho_0) \sim T^{\varepsilon}$ that is found below 2 K [3]. As pointed out by Rosch [7] the T dependence of ρ near an AFM QCP is strongly affected by impurity scattering. While $T^{3/2}$ is predicted for samples with short electronic mean free path, i.e. in the "dirty limit", a reduction of ρ_0 should result in a decrease of ε towards $\varepsilon = 1$ for very small but finite ρ_0. We indeed observe $\varepsilon = 1.5$ for $\rho_0 \geqslant 3$ μΩ cm and $1.3 \leqslant \varepsilon < 1.5$ for 0.17 μΩ cm $\leqslant \rho_0 < 3$ μΩ cm.

To summarize, incipient SC is found in the Ni-rich part of the homogeneity range of $CeNi_2Ge_2$. Ni-excess leads, similar to Cu-excess in $CeCu_2Si_2$, to a slight increase of the hybridization g.

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