First observation of superconductivity in $LaCu_6$ and possible applications

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Abstract

We have measured the ac susceptibility and resistivity of highly pure samples of the intermetallic compound $LaCu_6$ down to ultralow temperatures. We have prepared the samples by arc melting of stoichiometric amounts of 99.99% La and 99.9999% Cu in a water-cooled copper crucible under Ar protective atmosphere and analysed them by x-ray diffraction and SQUID magnetometry. At $T \leq T_c = 0.16K$ we observe a superconducting transition. Due to the manifold physical properties of isostructural $ReCu_6$ compounds (e.g. RE = Ce: heavy fermion system, RE = Pr: hyperfine enhanced nuclear spin system, RE = Nd: electronic antiferromagnet), numerous studies of interplay phenomena may become possible in the quasibinary compounds $RE_{1-x}La_xCu_6$, respectively.

Key words: superconductivity; interplay phenomena in superconductors; magnetic superconductors; heavy fermion superconductors

The isostructural intermetallic rare earth copper compounds of stoichiometry $RECu_6$ reveal an interesting variety of physical phenomena. $CeCu_6$ is a classical heavy fermion system which exhibits dynamic (e.g. Ref. 1) and static spin correlations (e.g. Ref. 2-4). $PrCu_6$ is a classical Van Vleck paramagnet and in respect of its nuclear magnetic properties, a so called hyperfine enhanced nuclear spin system which undergoes a spontaneous nuclear magnetic ordering transition at mK temperatures already (e.g. Ref. 5, 6). $NdCu_6$, finally, is well known as an electronic antiferromagnet. To this list of materials and their interesting properties, we add $LaCu_6$ which indeed is a simple Pauli paramagnet only, but has been firstly identified to be a superconductor, in the course of our investigation. This finding enables a long list of new possibilities to study multiple interplay phenomena of super- and normal conducting transport mechanisms as well as various magnetic properties in quasibinary, $RE_{1-x}La_xCu_6$, or in respect of quantum critical phenomena, even in quasiternary compounds, $RE_{1-x}La_xCu_{6-y}Au_y$.

In our work, we have prepared polycrystalline $LaCu_6$ by arc melting of about stoichiometric amounts of 99.99%pure La and 99.9999% pure Cu. In more detail, we have added 5 atom % of Cu to the stoichiometric ratio in order to compensate the higher volatility of Cu from the melt. The sample was molten in a water cooled copper crucible under 99.9999% Ar atmosphere at 0.5 bar pressure. For best homogeneity, the sample was several times turned and remolten.

In order to examine the sample quality, we have performed X-ray diffraction as well SQUID magnetization measurements. The x-ray diffraction data are in agreement with calculated diffraction patterns (see Fig.1). The small deviations of data from the calculation are very similar to those observed for a isostructural high quality $CeCu_6$ crystal, investigated before, and may mainly result from surface oxide contributions (additional intensity lines) and texture effects (quantitative differences of intensities). The use of a Cu-K_α anode in the diffractometer and the high Cu content in the material are responsible for the low intensity to back-

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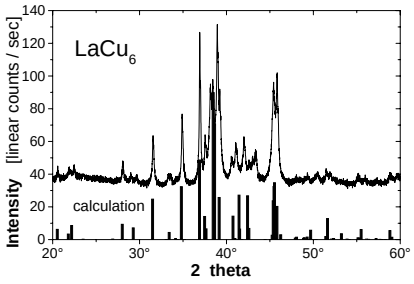

Fig. 1. X-ray powder diffraction data of our $LaCu_6$ compared to a calculation using the 'lazy pulverix' software [7].

ground ratio due to Cu x-ray fluorescence. The content of magnetic impurities in our $LaCu_6$ sample we have analysed by polarising the impurity moments in a field of 3.0 T at Kelvin temperatures and fitting the Brillouin function to the SQUID magnetization data given in Fig.2. We obtain average values of the concentration $x = (26 \pm 2)ppm$, of the single magnetic moments $\mu = (11.7 \pm 0.9)\mu_B$, and of the average spin quantum number $J = 2.5 \pm 0.6$ of the magnetic impurities, reflecting a distribution of magnetic rare earths in 99.99% La.

The ultralow temperature properties of $LaCu_6$ we have investigated in our nuclear demagnetization refrigerator [8]. In detail, we have measured the ac resistivity and ac susceptibility at a frequency of 16 Hz. In order to get a reasonable thermal coupling of the used bulk $LaCu_6$ sample to the thermal reservoir, it has been directly connected via a silver link to the nuclear cooling stage. Resistivity data have been taken by cooling down the cryostat from room temperature to a few mK. As it is typical for intermetallic compounds, we observe a rather small residual resistivity ratio $RRR \approx 11$ for our sample (see Fig.3). At about 0.27 K a small drop of the resistivity R from the normal residual resistivity RR as a possible precursor for superconductivity sets in. The reduction of R to 90% RR as a first significant drop, 50% RR, and 10% RR occurs at 160 mK, 120

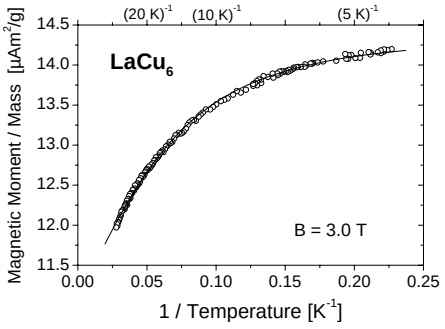

Fig. 2. Magnetic moment per mass of our $LaCu_6$ which has been exposed to a magnetic field of 3.0 T as function of inverse temperature at $(40K)^{-1} \leq T \leq (4.3K)^{-1}$. A fit of the Brillouin function (line) to the data points leads to the average values of the magnetic impurity quantities given in the text.

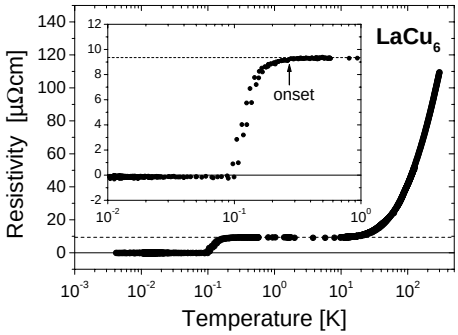

Fig. 3. Ac resistivity data of bulk $LaCu_6$ in the wide temperature range, $4mK \leq T \leq 300K$. The inset shows the superconduction transition in more detail. The onset of the broadened transition occurs at T = 0.27 K, already. The bulk $LaCu_6$ sample has been exposed to a static field $B_0 \leq 10\mu T$.

mK, and 100 mK, respectively. Below 100 mK the resistivity remains zero. The superconducting transition of our sample appeares to be clearly broadened. The susceptibility data (not shown here due to limited space) exhibit similar features. We suspect that the broadening is at least partly caused by the impurity content of magnetic rare earths, analysed above. Due to the quenched magnetic properties in $CeCu_6$ through the Kondo effect and in $PrCu_6$ through the singlet crystal field ground state we expect a much smaller influence of Ce and Pr doping on the superconducting transition in $LaCu_6$.

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References

[1] G. Aeppli et al., Phys. Rev. Lett. **57** (1986) 122

[2] C. Jin et al., Physica B **51** (1995) 12892

[3] T. Herrmannsdörfer et al., Physica B **284-288** (2000) 1263

[4] H. Tsujii et al., Phys. Rev. Lett. **84** (2000) 5407

[5] J. Babcock et al., Phys. Rev. Lett. **43** (1979) 380

[6] K. Akashi et al., Phys. Rev. Lett. **82** (1999) 1297

[7] http://crystsun1.unige.ch/ilicrx/lazy.html

[8] K. Gloos et al., J. Low Temp. Phys. **73** (1988) 101