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Ferromagnetism, superconductivity and quantum criticality in uranium intermetallics

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2. Experimental

2.1. Sample preparation

The samples used and described in this thesis were all prepared at the Van der Waals - Zeeman Institute (WZI) of the University of Amsterdam.

Polycrystalline samples were prepared in a home-made mono-arc furnace. The starting materials of purity of at least 3N were weighed according to their (nominal) compositions and melted together in a water-cooled copper crucible under high-purity argon atmosphere after pre-evacuation to 10^{-7} mbar. The samples were turned over and re-melted several times to attain good homogeneity. The mass loss observed was below 0.1% and could be neglected. A heat treatment is necessary to obtain better homogeneity for each sample and to reduce internal stress. The as-cast buttons were wrapped in Ta foil and annealed in water free quartz tubes under high vacuum for 10 days at 875 °C.

Single-crystalline samples were grown by Y. K. Huang. The single-crystalline rods were pulled from the melt using a modified Czochralski technique in a tri-arc furnace under a high-purity argon atmosphere. In the case of UCoGe an annealing procedure at high temperature (24 h at 1250 °C) then at lower temperature (21 days at 880 °C), like applied for URhGe [41], was employed to significantly improve the sample quality.

Samples for different experiments were mostly cut in a bar- or cube-shape by an AGIEPLUS spark-erosion machine with a precision position accuracy of 5 μm , after which the defected surface was removed by polishing.

2.2. Sample characterization

The crystallographic structures of the polycrystalline samples at room temperature were verified by a Philips APD-1700 diffractometer at the WZI, or a Bruker D3 Advance X-ray diffractometer at the Delft University of Technology, using Cu-K α radiation. The samples were powdered with Si used as a standard, and fixed in a random orientation by Apiezon grease and covered by Kapton foil to prevent contamination. The obtained powder XRD patterns were analyzed by means of a Rietveld refinement [42] procedure using X'pert HighScore Plus [43] or the GSAS programs [44] in order to determine the type of structure and the lattice parameters. The analyses showed no signs of impurity phase within a resolution of 5 vol.%.

Single-crystallinity was checked by means of X-ray back-scattering in the standard Laue geometry. The Cu-K α radiation generated with 40 mA at 20 kV was utilized as an incident beam and the reflections were recorded on a Polaroid photographic film. The Laue photo gives information on crystal quality over an area of about 1 mm². The program OrientExpress [45], which is able to generate the whole Laue pattern from a few main observed reflection spots, was used to orient the crystals.

The phase homogeneity and the stoichiometry of samples were thoroughly investigated with electron probe microanalysis (EPMA). These measurements have been done with the JEOL JXA-8621 equipment at the WZI. Small amounts (2 - 3%) of impurity phases were detected in the polycrystalline samples, whereas the single crystals turned out to be single phase.

2.3. Magnetization measurement

The temperature and magnetic field dependences of the magnetization were measured with a Quantum Design SQUID magnetometer MPMS-XL [46] at the WZI. The sensitivity of this equipment is as high as 10⁻⁸ emu. The temperature range is from 1.7 to 350 K and the magnetic field produced by the superconducting magnet can range from -5 to 5 T. A sample is inserted in a gelatin capsule and mounted in a transparent plastic straw with a diamagnetic contribution of the order of 10⁻⁵ emu in 1 T. After field cooling, the magnetization is detected by the magnetic flux change in the superconducting loop induced by the movement of the sample. The demagnetizing factor of our bar-shaped samples with

typical dimensions of $1 \times 1 \times 5 \text{ mm}^3$ is small ($N \approx 0.08$) [47].

High-field magnetization measurements were carried out in magnetic fields up to 52 T at 4.2 K by using the pulsed magnetic field facility at the Laboratoire National des Champs Magnétiques Pulsés (LNCMP) in Toulouse, France. The mass amounted to $\sim 20 \text{ mg}$ in the case of the cube-shaped single crystal, and to $\sim 40 \text{ mg}$ for the bar-shaped polycrystals.

2.4. Resistivity measurement

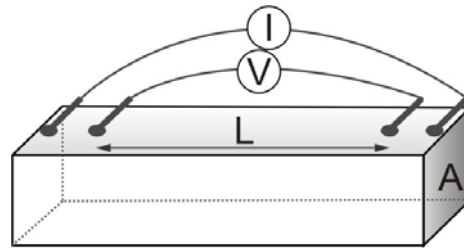


Figure 2.1 Contact geometry in resistivity measurements.

Measurements of the electrical resistance R were performed using a standard four-probe low frequency ac-technique available at the WZI. In this setup, a current I is passed through the sample by two outer leads, while the potential drops V across the sample is measured using two inner leads. The electrical contacts were made by using Cu wire ($30 \text{ }\mu\text{m}$ diameter) connected to the sample by silver paste. The contact geometry for the bar-shaped samples with a cross section $A \sim 1 \text{ mm}^2$ is shown in Fig. 2.1. The distance L between the voltage probes varied from 2 to 6 mm. The resistivity ρ and resistance R are related according to:

$$\rho = R \frac{A}{L} \quad (2.1)$$

The uncertainty in the determination of the geometrical factor A/L is about $\sim 10\%$, and is mainly due to the relatively large spread of the voltage contacts made by the silver paste. Also, since some of the samples were quite brittle, care should be taken when interpreting the absolute resistivity values. Cracks in the samples might lead to an effective cross section A_{eff} which is significantly smaller than the measured value, in which case the resistivity values calculated with help of Eq. 2.1 are a factor A/A_{eff} too large. However, the Residual Resistance Ratio ($RRR = \rho(300\text{K})/\rho_0$), which is a measure for the quality of the samples, is not affected by changes in the geometrical factor due to metallurgical problems.

The temperature dependence of the resistivity between 2 and 300 K was measured using an Oxford Instruments MagLab 2000 system operating at a frequency of 17 Hz and excitation currents of 5 - 10 mA while warming the samples at a heating rate of 0.4 K/min.

In the temperature range below 25 K, resistivity data were obtained using a Linear Research AC Bridge Resistance model LR700 operating at a frequency of 16 Hz and low excitation currents 10 - 100 μ A. Experiments were carried out in an Oxford Instruments HelioxVL 3 He system ($T_{\text{base}} = 250$ mK) [48] and an Oxford Instruments Kelvinox MX100 dilution refrigerator ($T_{\text{base}} = 20$ mK) [49]. The temperature of the sample is controlled by an Oxford LabVIEW program and read out from a RuO₂ thermometer mounted on the sample platform. These instruments are integrated with superconducting magnets providing magnetic fields up to 14 T in the Heliox and 18 T in the Kelvinox, respectively. This enabled to study the magnetic field dependence of the resistivity at low temperatures.

2.5. ac-susceptibility measurement

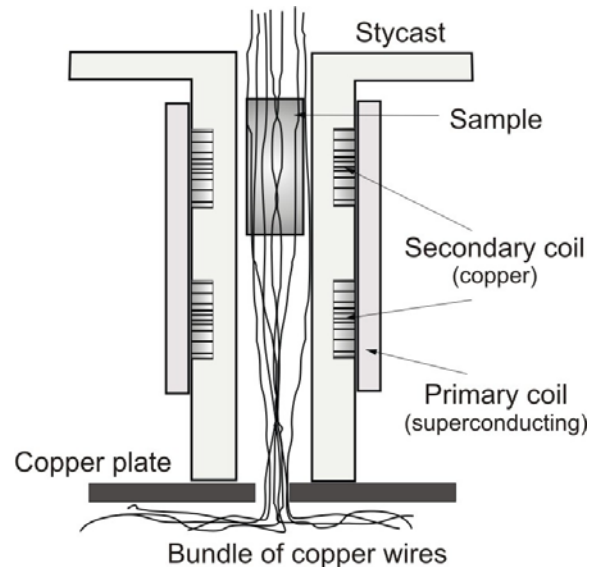


Figure 2.2 Schematic view of a mutual-inductance transformer for the ac-susceptibility measurement (from Ref.[51])

Ac-susceptibility measurements were used to detect the superconducting state and to determine the ordering temperatures of the magnetic compounds. A mutual-inductance transformer method is applied [50]. The primary coil is made of a superconducting wire (with a resistance of 32 k Ω at room temperature). Two secondary coils made of copper wire (with a resistance of 0.5 k Ω at room temperature) are wound in an opposite direction and

placed inside the primary coil as illustrated in Fig. 2.2 (taken from Ref.[51]). In this configuration, if the coils are connected then the out-put signal is equal to zero for perfectly balanced coils. The sample is wrapped in a bundle of copper wires that is thermally anchored to the cold plate and then inserted in the center of one of the secondary coils. The primary coil generates a magnetic field of 2.6 Oe at a current of 1 mA. The response of the sample to the generated field is picked up via the secondary coils. The signal is a direct measure of the ac-susceptibility.

The experiments were performed at low temperatures in the Heliox and Kelvinox systems following two methods:

- (i) the temperature- and field-dependence of the ac-susceptibility was measured at a fixed frequency of 16 Hz using the LR700 bridge with driving field of the order of 10^{-5} T.
- (ii) the temperature dependence of the zero-applied field susceptibility was measured at different frequencies between 35 Hz and 3.5 kHz using an EG&G 7265 DSP lock-in amplifier.

2.6. Specific-heat measurements

The specific heat was measured in a home-built set-up using a semi-adiabatic method employing a mechanical heat switch in a ^3He cryostat equipped with a 17 T superconducting magnet [52]. Electrical heat pulses of 15 to 30 seconds duration are applied to a sample holder, made of gold-plated cold-rolled silver. The temperature before and after the heat pulse is monitored by a so-called combination thermometer which exhibits a very limited field dependence [53]. The ^3He insert is a closed system, working with a room temperature gas storage vessel, and a cryopump for cooling the system down to 300 mK. The samples with mass of 3 - 4 g were clamped to the holder by a screw. The average accuracy of the measurement in the whole temperature range is 2%. The specific-heat measurements described in this thesis were performed in a temperature range between 0.3 and 20 K with different persistent applied magnetic fields ranging from 0 to 12 T.

In addition, the specific heat at lower temperatures 0.1 - 1 K was measured in a ^3He - ^4He dilution refrigerator at the University of Karlsruhe. This setup used a compensated-loss method with a weak thermal link and a sample with mass 0.1 g.

2.7. μ SR spectroscopy

μ SR stands for Muon Spin Rotation, Relaxation or Resonance and is a microscopic technique using the positive muon μ^+ as a probe. In recent years, μ SR has been widely applied to study the magnetic and superconductivity properties of f -electron materials exhibiting novel ground states (for review see [54,55]).

The μ SR experiments presented in this thesis were carried out using the μ^+ SR-dedicated beam line on the PSI-600MeV proton accelerator at the Swiss Muon Source of the Paul Scherrer Institute (PSI) in Villigen, Switzerland. A ^4He flow cryostat is used for measurements in the temperature range above 1.5 K at the General Purpose Spectrometer (GPS) [56]. For measurements below 1.5 K, an Oxford Instruments top-loading ^3He - ^4He dilution refrigerator ($T_{\text{base}} = 10$ mK) was used at the Low Temperature Facility (LTF) [57]. In the GPS the sample was mounted by Mylar tape on a fork-like holder, while in the LTF the sample was glued to a silver holder by General Electric varnish.