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### Ferromagnetism, superconductivity and quantum criticality in uranium intermetallics

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# 5. Ferromagnetic quantum critical point in single-crystalline URh<sub>0.62</sub>Ru<sub>0.38</sub>Ge

## 5.1. Introduction

In Chapter 4 we have reported the evolution of ferromagnetism of the polycrystalline URh<sub>1-x</sub>Ru<sub>x</sub>Ge series. Upon Ru doping ferromagnetism is smoothly suppressed and vanishes at the FM QCP  $x_{\text{cr}} = 0.38$ . Non-Fermi Liquid (NFL) behaviour is observed for samples with Ru concentration near  $x_{\text{cr}}$ . This offers a rare opportunity to study FM spin fluctuations near a ferromagnetic quantum critical point at ambient pressure. However, such a study preferably should be carried out on single-crystalline samples, as one expects a strong magneto-crystalline anisotropy for the orthorhombic TiNiSi structure [40]. Especially, inelastic neutron scattering studies to probe the critical fluctuations require large high-quality single-crystals.

In this chapter, we report the first investigations of a single-crystalline sample URh<sub>1-x</sub>Ru<sub>x</sub>Ge with  $x$  near the critical concentration  $x_{\text{cr}} \approx 0.38$ . Transport, magnetization and specific-heat measurements in high magnetic fields have been carried out. The data reveals a strong anisotropy of the easy-plane type, with the  $a$ -axis as the hard axis. Upon applying a magnetic field our sample is tuned away from the FM QCP and the Fermi liquid state is recovered.

## 5.2. Sample preparation and characterization

A polycrystalline batch with nominal composition U<sub>1.01</sub>Rh<sub>0.62</sub>Ru<sub>0.38</sub>Ge was prepared by

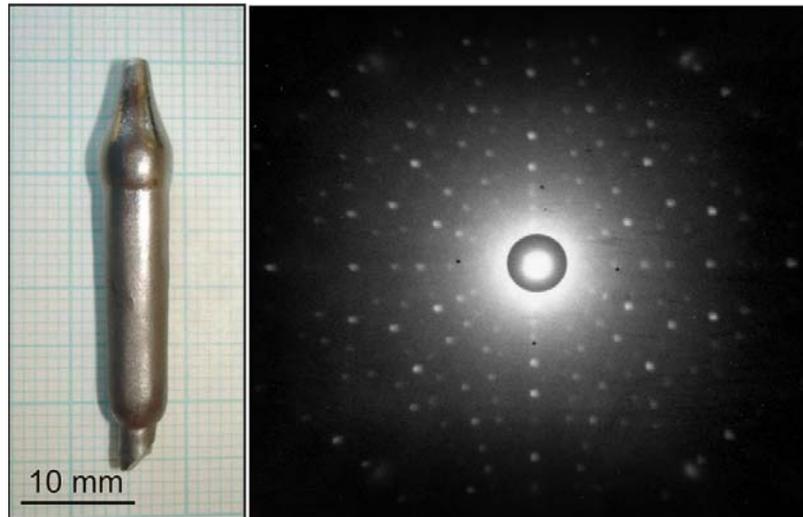


Figure 5.1 X-ray Laue picture (right panel) of as-grown single crystalline  $\text{URh}_{0.62}\text{Ru}_{0.38}\text{Ge}$  (left panel).

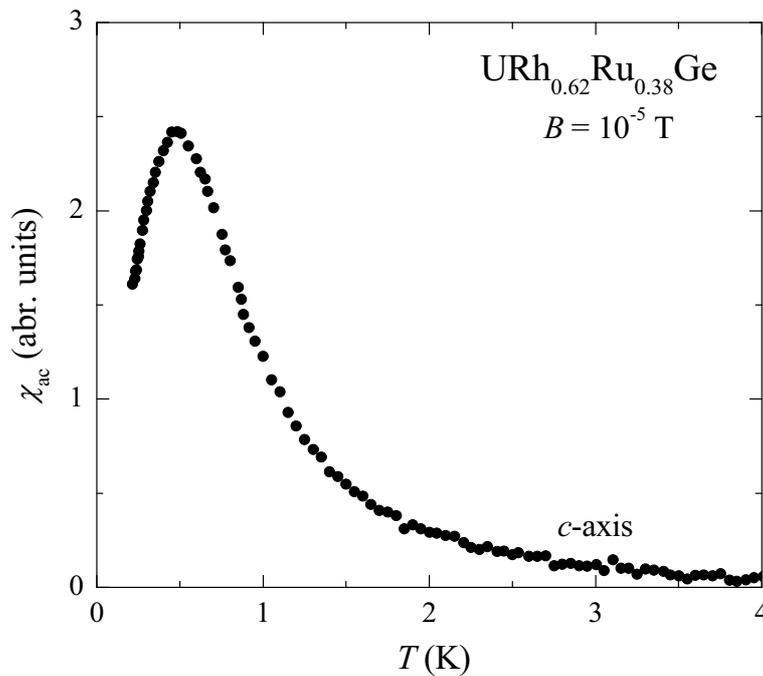


Figure 5.2 Temperature dependence of the ac-susceptibility for the  $\text{URh}_{0.62}\text{Ru}_{0.38}\text{Ge}$  crystal measured at a low frequency of 16 Hz and in a small driving field of  $\sim 10^{-5}$  T applied along the  $c$ -axis (data taken by A. Gasparini [145]).

arc-melting the constituents (natural U 3N, Rh 3N, Ru 3N and Ge 5N) in a water-cooled copper crucible under a high-purity argon atmosphere. The polycrystalline ingot was used as a seed for the single-crystal, grown by a modified Czochralski method in a tri-arc furnace under a high-purity argon atmosphere. No subsequent heat treatment was given to the single-crystal. In Fig. 5.1 we show a picture and Laue pattern of the single-crystal,

which is 5 - 6 mm in diameter and 35 mm in length. EPMA analysis confirmed that the crystal is single-phase and homogeneous. Single-crystallinity was checked by X-ray Laue backscattering. Samples for various measurements were cut by spark erosion.

Ac-susceptibility measurements carried out on a small piece of the crystal [145] show a weak ordering peak at a temperature around 0.4 K, see Fig. 5.2. This indicates that the single-crystal orders ferromagnetically at a Curie temperature  $T_C \approx 0.4$  K and that  $x < x_{cr}$ . When comparing with  $T_C(x)$  obtained for the polycrystalline samples (see Fig. 4.17), we conclude that  $T_C = 0.4$  K corresponds to  $x = 0.375$ . Thus the actual Ru content in the single-crystal appears to be slightly lower the nominal value  $x = 0.38$ . As mentioned in Section 4.2, we cannot determine the exact atomic ratio of Rh and Ru using the EPMA technique, and in the following we use the nominal value  $x = 0.38$ .

### 5.3. Electrical resistivity

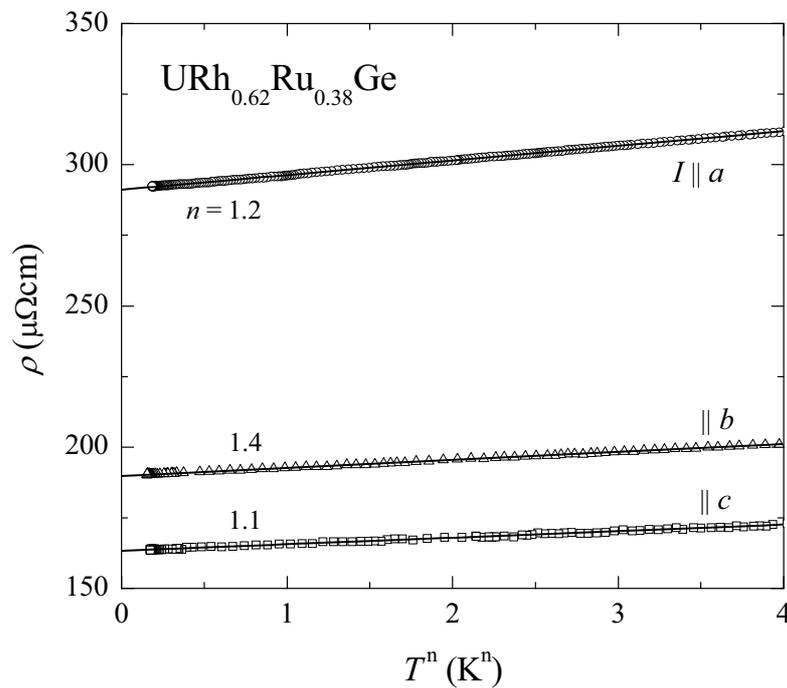


Figure 5.3 The resistivity of single-crystalline  $URh_{0.62}Ru_{0.38}Ge$  plotted versus  $T^n$  for a current along the principal axes, as indicated. The solid lines represent fits of the data to  $\rho \sim T^n$ .

In Fig. 5.3, we show the temperature variation of the resistivity of the  $URh_{0.62}Ru_{0.38}Ge$  single-crystal plotted as  $\rho$  versus  $T^n$  for the current  $I$  along the principal axes. The overall resistivity values for  $I \parallel a$  are about a factor 3/2 larger for  $I \parallel b$  and  $I \parallel c$ . The residual

resistance ratio's ( $RRR$ ) are quite low and amount to  $\sim 2$ . By fitting the data to  $\rho \sim T^n$  in the temperature range 0.25 - 4 K, we extract values for the exponent  $n$  of 1.2, 1.4 and 1.1 for  $I \parallel a$ ,  $b$  and  $c$ , respectively. These  $n$  exponents are close to the value  $n \approx 1.2$  obtained for polycrystalline URh<sub>1-x</sub>Ru<sub>x</sub>Ge with  $x \sim 0.38$  (see Fig. 4.12). We conclude the non-Fermi liquid (NFL) behaviour locates our sample close to the FM QCP. In Table 5.1, we summarize the fit-parameters from the transport data.

For polycrystalline URh<sub>0.62</sub>Ru<sub>0.38</sub>Ge, resistivity measurements in magnetic fields demonstrated that the FL state is recovered upon field application of the magnetic field. The effect of a magnetic field on the resistivity of the single-crystalline samples has not been investigated yet.

*Table 5.1* The parameters  $\rho_0$ ,  $n$  and  $A$  obtained by fitting the resistivity to  $\rho = \rho_0 + AT^n$  (Eq. 4.5) for the URh<sub>0.62</sub>Ru<sub>0.38</sub>Ge crystal.

$I$ direction	$\rho_0$ ( $\mu\Omega\text{cm}$ )	$n$	$A$ ( $\mu\Omega\text{cm}/\text{K}^n$ )
$a$ -axis	291	1.2	5.19
$b$ -axis	190	1.4	2.80
$c$ -axis	163	1.1	2.31

#### 5.4. Magnetic properties

The field dependence of the magnetization of the URh<sub>0.62</sub>Ru<sub>0.38</sub>Ge single-crystal, measured in magnetic fields up to 50 T applied along the principal axes at  $T = 4.2$  K, is shown in Fig. 5.4. The magnetization  $M$  is strongly anisotropic. The induced moment initially grows fastest for  $B$  along the  $c$ -axis, but for  $B > 27$  T  $M_b > M_c$ . The magnetic anisotropy is of the easy-plane type, with the  $a$ -axis being the hard axis. For  $B \parallel a$ , the magnetization linearly increases up to the highest field. The anisotropy in the magnetization of doped URhGe mimics the anisotropy observed for pure URhGe, where a spin reorientation process takes place in the  $bc$ -plane for a field of 12 T directed along the  $b$ -axis. The induced moment in the  $bc$ -plane near 50 T amount to  $0.6 \mu_B$ , which is still much smaller than the saturation values  $M_S = 3.58$  or  $3.62 \mu_B$  for the  $5f^2$  and  $5f^3$  configurations. We did not observe a positive curvature of  $dM/dB$  that could hint at a spin-reorientation, or a re-entrant FM transition, predicted for a compound located in the vicinity of a ferromagnetic quantum critical end-point [89].

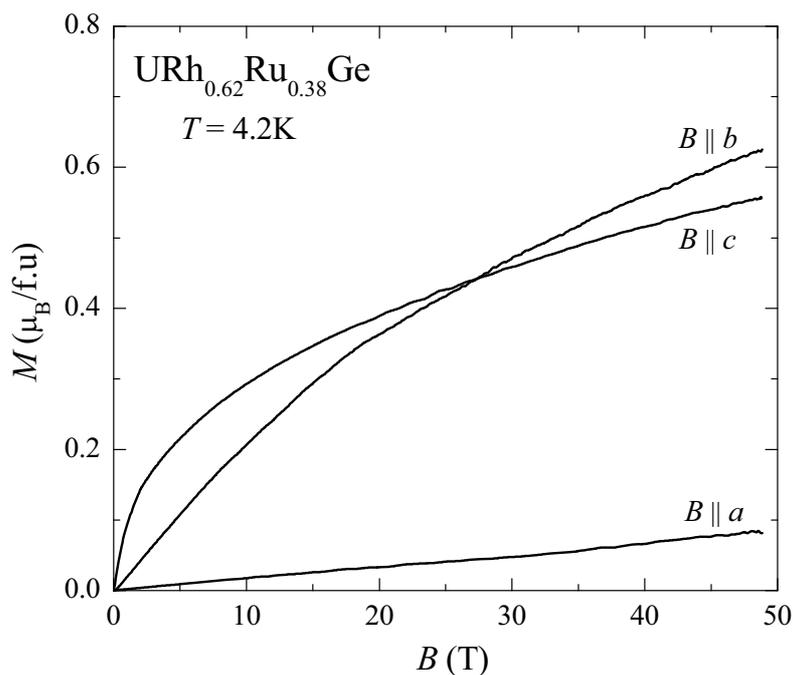


Figure 5.4 The high-field magnetization of single-crystalline  $\text{URh}_{0.62}\text{Ru}_{0.38}\text{Ge}$  measured in a field up to 50 T applied along the principal axes as indicated, at  $T = 4.2$  K.

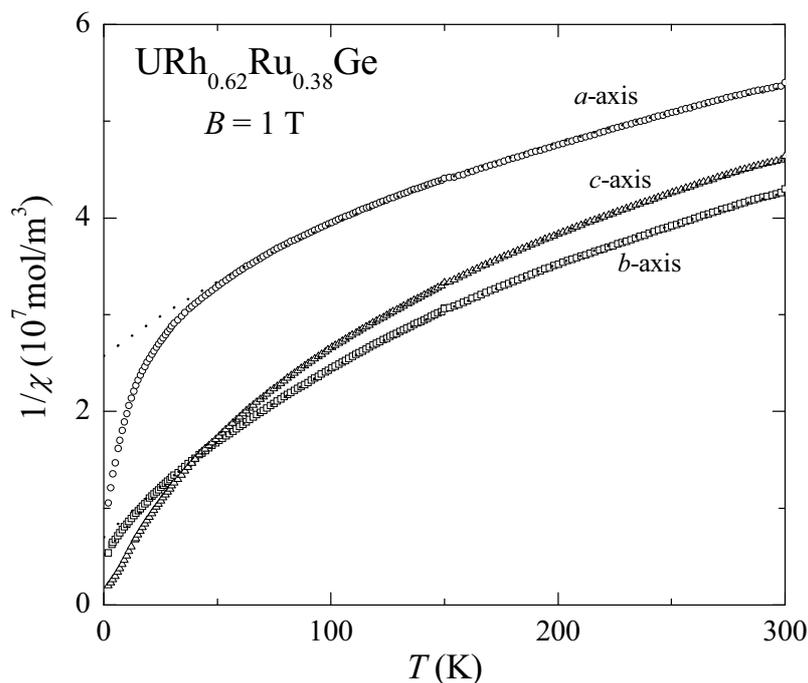


Figure 5.5 Temperature variation of the reciprocal susceptibility of the  $\text{URh}_{0.62}\text{Ru}_{0.38}\text{Ge}$  single-crystal measured in a field of 1 T applied along the principal axes. The dotted lines are the best fits to a MCW law in the temperature range 50 - 300 K.

The reciprocal susceptibility of the single-crystalline  $\text{URh}_{0.62}\text{Ru}_{0.38}\text{Ge}$  sample measured in a field  $B = 1$  T applied along the  $a$ -,  $b$ - and  $c$ -axis in the temperature range 2 - 300 K is shown in Fig. 5.5. Above 50 K the magnetic susceptibility follows a MCW law with

$\chi_0 \sim 10^{-8} \text{ m}^3/\text{mol}$ . The data confirm a magnetic anisotropy, of the easy-plane type with the hard-magnetization direction along the  $a$ -axis. This is also reflected in the paramagnetic Curie temperature  $\theta_a \gg \theta_b, \theta_c$ . The values of the effective moment  $p_{\text{eff}}$  listed in Table 5.2 indicate that the largest moment is induced for  $B \parallel b$  in the paramagnetic region ( $T > 50 \text{ K}$ ). This is in good agreement with the susceptibility data reported on single-crystalline URhGe [119].

Table 5.2 The effective moment  $p_{\text{eff}}$  and the paramagnetic Curie temperature  $\theta$  of the URh<sub>0.62</sub>Ru<sub>0.38</sub>Ge single-crystal as deduced from magnetization measurements.

$B$ direction	$p_{\text{eff}} (\mu_{\text{B}}/\text{f.u.})$	$\theta (\text{K})$
$a$ -axis	1.32	-101.5
$b$ -axis	1.53	-28.1
$c$ -axis	1.38	-16.6

## 5.5. Specific heat

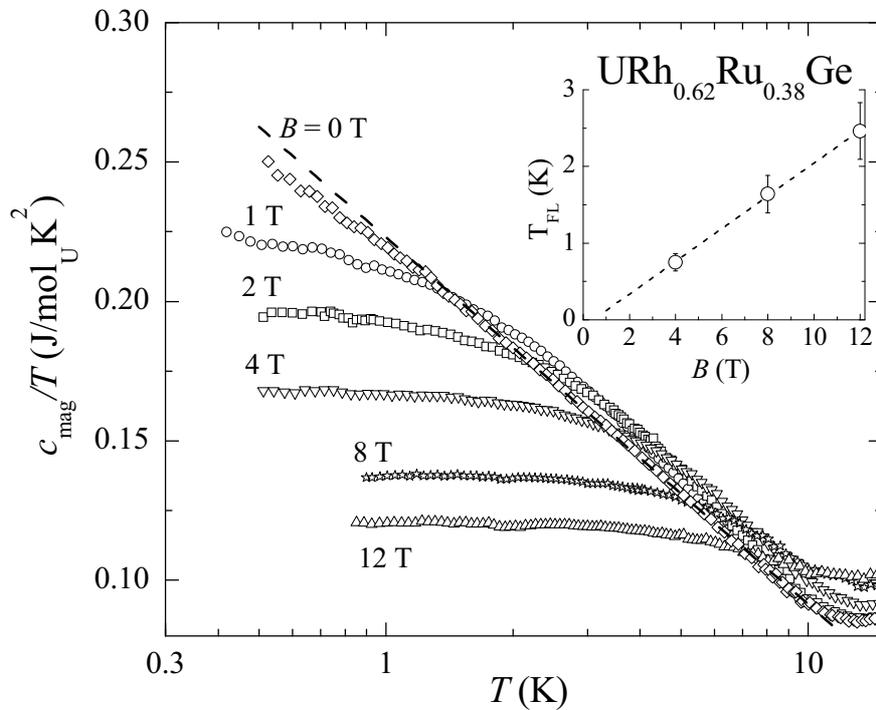


Figure 5.6  $f$ -electron specific heat of single-crystalline URh<sub>0.62</sub>Ru<sub>0.38</sub>Ge plotted as  $c_{\text{mag}}/T$  versus  $T$  (on a logarithmic scale) in magnetic fields of 0, 1, 2, 4, 8 and 12 T applied along the easy axis  $c$ . The dashed line represents a fit  $c_{\text{mag}}/T = -b \ln(T/T_0)$  (Eq. 4.9) with  $b = 0.057 \text{ J/molK}^2$  and the scaling temperature  $T_0 = 49.5 \text{ K}$  for the data measured in zero field. *Inset*: the field-dependence of  $T_{\text{FL}}$ .

Specific heat measurements on the  $URh_{0.62}Ru_{0.38}Ge$  crystal are performed in magnetic fields up to 12 T in the temperature interval 0.5 - 20 K. The magnetic field was directed along the  $c$ -axis, *i.e.* the easy direction for magnetization, in order to induce the largest effect. The lattice contribution to the specific heat,  $c_{\text{lat}} = \beta T^3$ , is obtained by fitting the data measured in zero field in the temperature range  $10 \text{ K} < T < 20 \text{ K}$ . We find  $\beta = 0.60 \times 10^{-3} \text{ J/molK}^4$  and  $\theta_D \approx 210 \text{ K}$  which are the same values as obtained for the polycrystalline samples (see Section 4.5).

In Fig. 5.6, we show the  $f$ -electron specific heat divided by temperature  $c_{\text{mag}}/T$ , obtained after subtracting the lattice contribution, in different fields. Here the data are plotted on a logarithmic temperature scale.

The zero-field data show pronounced NFL behaviour. Below 10 K  $c_{\text{mag}}/T$  grows in a (quasi)logarithmic fashion down to 2 K:  $c_{\text{mag}}/T = -b \ln(T/T_0)$  with  $b = 0.057 \text{ J/molK}^2$  and the scaling temperature  $T_0 = 49.5 \text{ K}$ . The values for  $b$  and  $T_0$  are almost identical to the values obtained for the polycrystalline sample at the critical concentration  $x_{\text{cr}} = 0.38$ . However, below 2 K the data deviate from the  $\ln T$  behaviour, because for the single-crystal  $x < x_{\text{cr}}$  and magnetic order sets in at  $\sim 0.4 \text{ K}$  (see Fig. 5.2)

In a magnetic field applied along the  $c$ -axis,  $c/T$  is suppressed and levels off towards a constant value for  $T \rightarrow 0$ . This shows the FL state is recovered in a field. The characteristic temperature  $T_{\text{FL}}$ , at which  $c/T$  becomes constant, increases with magnetic field, as shown in the inset of Fig. 5.6. By linearly fitting the  $T_{\text{FL}}(B)$  data, we extract a small value  $T_{\text{FL}} \approx 0.2 \text{ K}$  when  $B \rightarrow 0$ . This reveals the single-crystal is located close to the QCP. In the temperature range  $T > 10 \text{ K}$  the magnetic contribution to the specific heat increases with applied fields, see Fig. 5.6. This shows entropy is transferred from low to high temperatures.

The  $c/T$  values measured at 0.5 K decrease gradually with magnetic fields.  $c/T(B)$  follows an exponential decay function [119] (see the left panel of Fig. 5.7)

$$c/T|_{0.5\text{K}}(H) = c/T|_{0.5\text{K}}(\infty) + A(1 - e^{-\mu_0 H / \Delta_B}) \quad (5.1)$$

Here  $c/T|_{0.5\text{K}}(\infty) = 0.113 \text{ J/molK}^2$  represents the value of  $c/T|_{0.5\text{K}}$  for  $B \rightarrow \infty$ ,  $\Delta_B = 4.23 \text{ T}$  describes how fast  $c/T|_{0.5\text{K}}$  decays with increasing field, and  $A = 0.136 \text{ J/molK}^2$  is a fitting parameter.

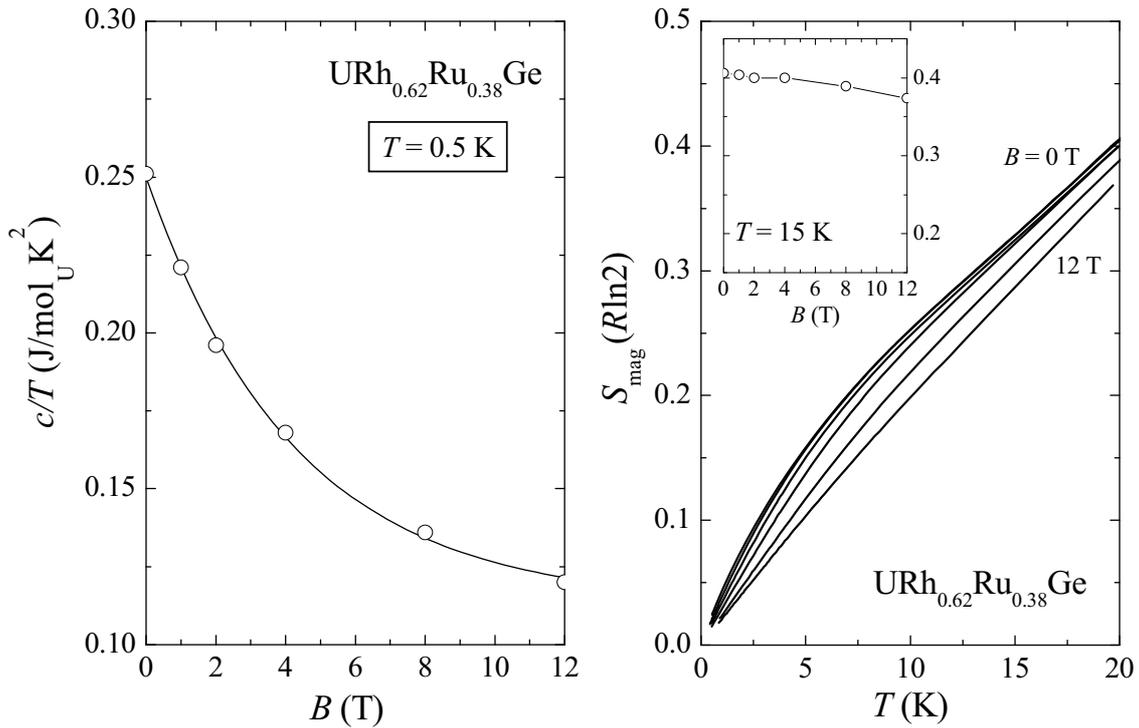


Figure 5.7 Left panel: The  $\gamma$  value ( $c/T$  value at 0.5 K or extrapolated for  $B = 8$  and 12 T) as a function of magnetic field  $B$ . Right panel: The magnetic entropy  $S_{\text{mag}}$  (in units of  $R \ln 2$ ) for the  $\text{URh}_{0.62}\text{Ru}_{0.38}\text{Ge}$  single-crystal. The applied fields are (from top to bottom)  $B = 0, 1, 2, 4, 8$  and 12 T. Inset: The field-dependence of  $S_{\text{mag}}$  at a fixed temperature of 15 K.

The magnetic entropy  $S_{\text{mag}}$ , which is calculated by integrating  $c_{\text{mag}}/T$  versus  $T$  between 0.5 and 15 K is shown in the right panel of Fig. 5.7. In zero field,  $S_{\text{mag}}$  is about  $0.33R \ln 2$  which is in good agreement with the value obtained on the polycrystalline sample with  $x = 0.38$ . Upon applying a magnetic field,  $S_{\text{mag}}$  slightly decreases to a value  $0.29R \ln 2$  for  $B = 12 \text{ T}$ .

## 5.6. Discussion

The transport and magnetization data of single-crystalline  $\text{URh}_{0.62}\text{Ru}_{0.38}\text{Ge}$  reveal strong magnetocrystalline anisotropy of the easy-plane type, with the hard-magnetization direction along the  $a$ -axis. This anisotropy reflects the one in pure  $\text{URhGe}$ .  $\text{URhGe}$  is a uniaxial ferromagnet with the magnetic moment along the orthorhombic  $c$ -axis [36]. However, upon application of a magnetic field along the  $b$ -axis the moment rotates in the  $bc$ -plane toward the  $b$ -axis, while the  $a$ -axis is the hard axis [38]. The strong magnetocrystalline anisotropy stresses the need to carry out all subsequent experiments, like neutron scattering to probe the anisotropic spin-fluctuation spectrum, on single-crystalline samples.

Upon application of a magnetic field  $B \parallel c$ , the FL state is recovered. Evidence for this is provided by the suppression of the specific heat, notably the levelling off towards constant values of  $c/T$  for  $T \rightarrow 0$ . A similar behaviour has been observed in other NFL doped-systems [146-148]. However, the only other FM case is the  $CePd_{1-x}Rh_x$  series [148]. Experiments on polycrystalline samples show that  $CePd_{1-x}Rh_x$  exhibits a “smeared” FM QCP at  $x_{cr} = 0.87$  [27]. The NFL behaviour observed near  $x_{cr} = 0.87$  is then a consequence of the distribution of single-ion Kondo temperatures, and can be explained by the Kondo disorder model [149]. Also, the  $c_{mag}/T$  versus  $T$  data of  $CePd_{1-x}Rh_x$  for  $x \sim x_{cr}$  develop a broad maximum at a characteristic temperature  $T_{max}$  upon applying a magnetic field [148].  $T_{max}$  increases linearly with increasing magnetic field and attains a value of  $\sim 1$  K for a field  $B = 3$  T. The field-induced broad maximum is tentatively attributed to a Schottky anomaly connected with the Zeeman splitting of the partially Kondo-screened crystal-electric-field ground-state doublet in the external magnetic field [150]. A similar phenomenon, *i.e.*, a maximum in  $c_{mag}/T$  versus  $T$ , is not observed in the specific heat data of the  $URh_{0.62}Ru_{0.38}Ge$  single-crystal:  $c_{mag}/T$  tends to level off and saturates in the limit  $T \rightarrow 0$  for fields up to 12 T. The difference in behaviour can be attributed to the very different scaling temperatures  $T_0$  deduced from the  $c_{mag}/T = -b \ln(T/T_0)$  behaviour. For  $URh_{0.62}Ru_{0.38}Ge$   $T_0 \approx 50$  K, while for  $CePd_{0.13}Rh_{0.87}$   $T_0 \approx 5.5$  K. This shows the NFL behaviour is more robust in  $URh_{0.62}Ru_{0.38}Ge$  and fields much larger than 12 T are needed to resolve the Zeeman splitting.

In conclusion, we have presented the first study of the FM QCP in a single crystal of  $URh_{0.62}Ru_{0.38}Ge$ . The results show a strong magnetocrystalline anisotropy of the easy- $(bc)$  plane type, with the  $a$ -axis as the hard-magnetization direction. Specific-heat data in a magnetic field ( $B_{max} = 12$  T) applied along the  $c$ -axis show that  $URh_{0.62}Ru_{0.38}Ge$  can be tuned away from the FM QCP and that the Fermi liquid state is recovered. In order to learn more about the anisotropic nature of the break-down of the NFL behaviour near the FM QCP further experiments are required, like transport measurement for various directions of the magnetic fields and specific-heat measurements for  $B \parallel a$  and  $B \parallel b$ . Finally, we recall that  $URh_{0.62}Ru_{0.38}Ge$  is one of the scarce materials at a FM QCP at ambient pressure, and thus offers a unique testing ground for the study of critical ferromagnetic spin fluctuations by microscopic techniques, like inelastic neutron scattering or NMR.