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Short-range magnetic ordering in $URh_{0.7}Ru_{0.3}Ge$

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Abstract

The magnetic properties of $URh_{0.7}Ru_{0.3}Ge$ were investigated by means of ac susceptibility, dc magnetization, specific heat and electrical resistivity measurements. We observed a maximum of the ac susceptibility at $T_{max} = 2.9$ K and its frequency dependence, but no spontaneous magnetization at 2 K. An alternative scenario for the observed feature, for example, spin-glass freezing, is not confirmed by dc field dependences of the ac susceptibility and dc magnetization. The data from specific heat and electrical resistivity studies down to 2 K do not suggest long-range magnetic ordering, and point to spin-fluctuation behaviour of the alloy. We have estimated a spin-fluctuation temperature of ∼27 K and Stoner enhancement factor of ∼490. The experimental data are discussed in the framework of spin-fluctuation theories.

(Some figures in this article are in colour only in the electronic version)

1. Introduction

The striking behaviour of non-Fermi liquid (NFL) systems, in particular, aspects related to the underlying microscopic mechanism, have received much attention in recent years [1]. In fact, a number of f-electron systems showing properties that deviate from the Landau–Fermi theory have been discovered, and for those several theoretical scenarios have been proposed. Basically, there exist at least three models accounting for the NFL behaviour of intermetallics; they are based on either single-ion physics of noninteracting local moments [2], on a magnetic instability of systems closing to quantum critical points (QCPs) [3–6] or on crystallographic disorder [7, 8]. However, the mechanism of the formation of the NFL state is not very clear, especially in the case of disordered systems, because the disorder can yield the spin-glass phase in addition to the NFL behaviour around the QCP [9, 10].

Recently, Sakarya *et al* reported the magnetic phase diagram of the solid solutions $URh_{1-x}Ru_xGe$ [11]. The magnetic ground state of parent compounds URhGe and URuGe was reported many years ago [12–15]. The first compound is an itinerant ferromagnet with the Curie temperature of 9.5 K, while the latter compound is a paramagnet showing spin-fluctuation behaviour. According to Sakarya *et al*, Ru substitution causes the disappearance of magnetic order at a composition of $x \sim 0.38$ [11]. This finding may generate a large interest because

of the opportunity for investigations of QCPs in this system. In order to look for QCPs we have reinvestigated the magnetic phase diagram of these alloys. The preliminary results of our investigations ([16] and [17]) have recently been presented at conferences. In short, our data indicated ferromagnetic ordering for $x < 0.3$, short-range magnetic order for alloys with $x = 0.3$–0.35 and non-Fermi-liquid (NFL) behaviour down to 2 K in alloys close to $x = 0.4$. In the context of a close relationship between the spin-glass phase and the NFL state, the lack of a spin-glass feature in alloys with $x = 0.3$–0.35 is rather interesting. Therefore, in the present work we report the results of ac magnetic susceptibility, dc magnetization, specific heat and electrical resistivity measurements for the alloy with $x = 0.3$ in more detail. We argue that neither long-range magnetic order nor spin-glass freezing has taken place. Instead, there are several evidences for a spin-fluctuation feature in the investigated alloy.

2. Experimental details

The sample studied in this work was prepared by arc melting stoichiometric amounts of the high-quality metals (U: 99.8%, Rh and Ru: 99.97% and Ge: 99.999%) under a pure argon atmosphere. Details of the synthesis procedure and quality checking are given elsewhere [16]. The ac magnetic susceptibility χ_{ac} measurements were carried out in the frequency range ω of 10–10 000 Hz, utilizing an Oxford Instruments Maglab susceptometer. We have also measured the ac susceptibility at 100 Hz in several dc magnetic fields up to $\mu_0 H = 5$ T. The dc magnetization (M) was carried out using a Quantum Design magnetometer (MPMS-5) in fields up to 5 T. The measurements were done under zero-field-cooling (ZFC) and field-cooling (FC) conditions on both powdered and bulk samples. The specific heat data C_p were collected using the thermal relaxation method in a Quantum Design PPMS, installed at the Max Planck Institute for Chemical Physics of Solids in Dresden. The dc electrical resistivity was measured using the conventional dc four-probe method. The sample was rectangular, with typical dimensions 0.5 mm × 0.5 mm × 5 mm. The voltage and current leads were made by Au wire, attached to the sample using a silver paste.

3. Results

For the investigated $URh_{0.7}Ru_{0.3}Ge$ alloy, the ac magnetic susceptibility does show clearly a broad anomaly at low temperatures in both components, real χ' and imaginary χ''. The existence of such χ_{ac} maxima suggests either a spin-glass-like or nonmagnetic state with some short-range ferromagnetic interactions. In figure 1 we display the susceptibility calculated as $\chi_{ac}(T) = \sqrt{\chi'^2 + \chi''^2}$, measured at several frequencies. The applied frequency has two pronounced effects on the $\chi_{ac}(T)$ curves. Clearly, with increasing frequencies the position of the susceptibility maximum T_{max} is shifted to higher temperatures, and the susceptibility amplitude at T_{max} decreases. These effects indicate a slowing down of fluctuations of the magnetic uranium moments, resembling very much that of classical spin-glass systems [18]. An attempt to find the relationship between T_{max} and ω reveals that T_{max} increases with ω as $T_{max} \propto \omega^n$, where $n = 0.022$ (see the inset of figure 1). For spin-glasses, the fractional relative change in the freezing temperature T_f per decade change in frequency, $\Delta T_f/(T_f \Delta \log \omega)$, has usually been taken as a characteristic parameter [19]. This ratio has experimentally found to locate between 6×10^{-3} (in diluted alloys like Cu Mn, Ag Mn [20, 21]) and 6×10^{-2} (in $La_{1-x}Gd_xAl_2$ [22]). The experimental data for $URh_{0.7}Ru_{0.3}Ge$ between 10 and 10 000 Hz may allow one to estimate $\Delta T_f/(T_f \Delta \log \omega)$ to be ∼0.05. A strong frequency dependence of T_{max} in the sample studied prompts us to suspect that the microscopic magnetic entities in the compound are probably not single uranium moments, but rather clusters of ferromagnetically

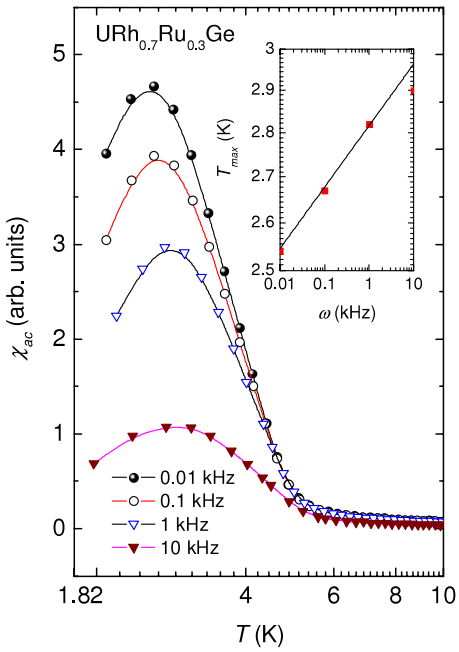

Figure 1. Temperature dependence of the ac susceptibility χ_{ac} of $URh_{0.7}Ru_{0.3}Ge$ measured at different frequencies. The inset shows a power relation between the position of the ac susceptibility maximum and the applied frequencies.

coupled uranium moments. A larger ratio $\Delta T_f/(T_f \Delta \log \omega) = 0.12$ was found in UCu_4Pd by Scheidt *et al* [23]. These authors have interpreted this as super-paramagnetism associated with spin clusters.

To further understand the behaviour observed in this system we measured the ac susceptibility at several dc magnetic fields up to 5 T. Actually, the experimental data provide arguments against spin-glass behaviour. Unlike the spin-glasses, where the magnetic fields shift T_{max} down to lower temperatures, we observe a quite opposite field dependence of T_{max} (figure 2); for example, as the strength of the applied field increases the position of the maximum is shifted to higher temperatures. Consequently, the classical Almeida–Thouless line $H \sim (1 - T_{max,H}/T_{max,0})^{3/2}$ [24], which describes the characteristics of spin-glasses in the mean-field framework, fails in the case of $URh_{0.7}Ru_{0.3}Ge$. However, one tries to describe the T_{max} versus H dependence utilizing the static scaling law, which was originally used for the second-order phase transition of a ferromagnet. According to the scaling hypothesis [25], the spontaneous magnetization M and susceptibility around T_C are governed by critical exponents β and γ, respectively, through the relations $M \propto (-\varepsilon)^{\beta}$ for $\varepsilon < 0$ and $\chi^{-1} \propto (\varepsilon)^{\gamma}$ for $\varepsilon > 0$, where $\varepsilon = \frac{T - T_C}{T_C}$. The field dependence of T_C is given by the law $\frac{T_C - T_{C,0}}{T_{C,0}} \propto H^{1/(\gamma+\beta)}$. Assuming T_{max} to be a crossover transition temperature of a short-range magnetic order, we may fit the data to the relation $\frac{T_{max} - T_{max,0}}{T_{max,0}} \propto H^{1/(\gamma+\beta)}$. For $URh_{0.7}Ru_{0.3}Ge$, the fitting of the data yields $\gamma + \beta = 1.49$ (see the inset of figure 2), which is comparable with that derived from the mean-field theory ($\gamma = 1$ and $\beta = 0.5$ [25]), but distinctly smaller than that predicted by either the three-dimensional Ising model ($\gamma = 1.24$, $\beta = 0.325$) or the Heisenberg model for magnetic systems ($\gamma = 1.336$, $\beta = 0.365$) [25]. Thus, there is no support for long-range ferromagnetic order due to localized moments in $URh_{0.7}Ru_{0.3}Ge$.

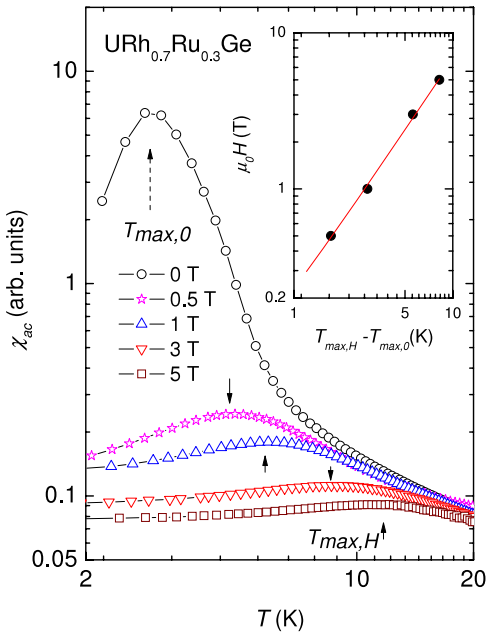

Figure 2. Temperature dependence of the ac susceptibility χ_{ac} of $URh_{0.7}Ru_{0.3}Ge$ measured at several magnetic fields. The inset shows a power relation between applied field and the position of the ac susceptibility maximum.

In figure 3, we show the temperature dependence of the dc magnetic susceptibility of $URh_{0.7}Ru_{0.3}Ge$, defined as $\chi_{dc} = M/H$, measured at 0.05, 0.5 and 2.5 T. An important observation emerging from the data is a strong temperature and field dependence of the susceptibility below 10 K. It leads to an upturn of the susceptibility with an inflection point T_{inf}, indicated by the arrow for $\chi_{dc}(T)$ at 0.05 T. This behaviour may be ascribed to the existence of short-range ferromagnetic interactions in the alloy. Consistently, we found that applied fields suppress the magnitude of $\chi_{dc}(T)$ and shift T_{inf} to higher temperatures. Since the system is not magnetically ordered we may apply the Stoner theory for nearly ferromagnetic metals [27, 28]. The theory predicted that the susceptibility of such systems at low temperatures should be quadratically dependent on temperature. Using the equation

$$\chi(T) = \chi(0) - aT^2, \tag{1}$$

we analysed the experimental data between 1.9 and 3.4 K with $\chi(0) = 1.01(1)$ cm^3 mol^{-1} and $a = 0.058(1)$ cm^3 mol^{-1} K^{-2}. The sizeable $\chi(0)$ value can be understood in terms of a large value of the Stoner enhancement factor S, which is given by the equation

$$\chi(0) = S\mu_B^2 N_0(\varepsilon_F), \tag{2}$$

where $N_0(\varepsilon_F)$ is the bare electron density of states at the Fermi level ε_F, and it can be estimated from the electronic specific heat of the bare electrons γ_0 through

$$N_0(\varepsilon_F) = \frac{3\gamma_0}{\pi^2 k_B^2}. \tag{3}$$

In the equations above, μ_B is the Bohr magneton and k_B is the Boltzmann constant.

In order to explore the possible irreversibility effect of the susceptibility, which is one of the basic characteristics of spin-glasses [19], one must compare the FC and ZFC susceptibility

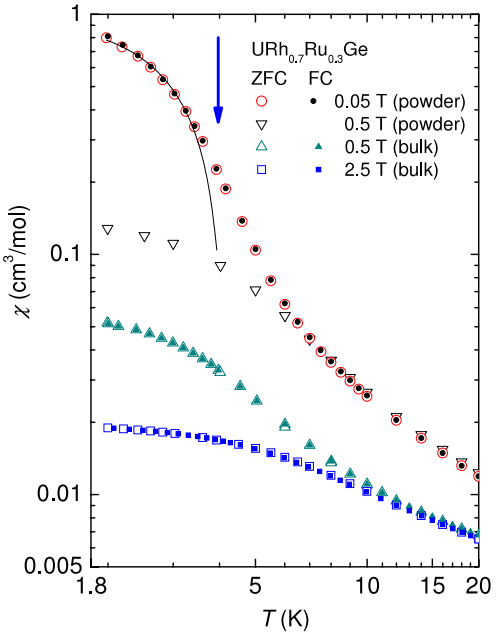

Figure 3. Temperature dependence of the dc susceptibility χ of $URh_{0.7}Ru_{0.3}Ge$ measured at several magnetic fields up to 2.5 T. The solid line is the fit to equation (1).

data. For $URh_{0.7}Ru_{0.3}Ge$, the FC and ZFC susceptibility curves collected in fields up to 2.5 T do not reveal any difference. Thus, the absence of any thermal irreversibility of the susceptibility implies that the anomaly observed at T_{inf} cannot be interpreted in terms of spin-glass freezing. We note also that the values of the susceptibility measured on powdered samples are larger than those obtained on bulk ones. The former data are the response of the system to magnetic fields, in which magnetic particles basically are free to be oriented. Thus, the results are believed to be close to that of an easy magnetization direction. The data obtained on bulk samples are simply those of polycrystalline material. The different behaviour of the powdered and bulk susceptibilities may be assigned to a strong magnetocrystalline anisotropy of the compound studied.

To follow the variation of the effective moment with temperature, and to access the information on the type of magnetic coupling between the uranium ions, we display in figure 4 the temperature dependence of the product χT of $URh_{0.7}Ru_{0.3}Ge$ at 0.05 and 0.5 T. In the case of lack of magnetic interaction, the Curie law predicts the product χT to be temperature independent. As a magnetic interaction is introduced, the product χT, according to the Curie–Weiss law, is then expected to change smoothly with temperature, showing either an upward or downward trend, depending on either ferromagnetic or antiferromagnetic interaction, respectively. For $URh_{0.7}Ru_{0.3}Ge$, however, the χT curves at low temperatures reveal a broad maximum, which certainly is a result of the competition between ferromagnetic and antiferromagnetic exchange interactions in the alloy. Inspecting the temperature dependence of the product χT, which starts to increase below 20 K, we may suggest that the fluctuations of the ferromagnetic entities just begin to give a rise to a χT upturn. With increasing magnetic field strength the magnitude of the maximum decreases remarkably, indicating a suppression of the fluctuations by the fields.

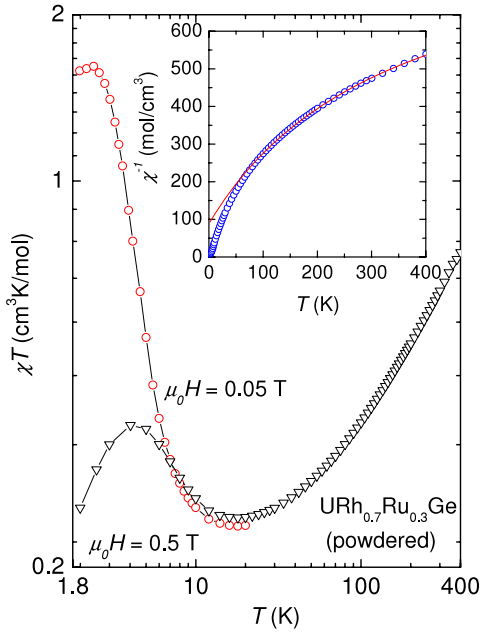

Figure 4. The product χT of $URh_{0.7}Ru_{0.3}Ge$ measured at 0.05 and 0.5 T. The inset shows the fit of the data to the modified Curie–Weiss law.

The presence of antiferromagnetic interactions in $URh_{0.7}Ru_{0.3}Ge$ may be confirmed by a large and negative paramagnetic Curie–Weiss temperature Θ_p. From a fitting of the susceptibility data for $T > 100$ K (see the inset of figure 4 to a modified Curie–Weiss law, $\chi = N_A\mu_B^2\mu_{eff}^2/3k_B(T - \Theta_p) + \chi_0$, where N_A is the Avogadro number, we obtained $\Theta_p = -32$ (1) K, $\chi_0 = 1.12 \times 10^{-3}$ cm^3 mol^{-1} and effective moment $\mu_{eff} = 1.63(1)\mu_B$. The independent temperature susceptibility, χ_0, denotes the Pauli susceptibility and the core diamagnetism as well as some Van Vleck paramagnetism. The obtained value of μ_{eff} is comparable with that found in URhGe, $\sim 1.8\mu_B$ [12, 26], and is considerably smaller than that expected for free U^{3+} ($3.62\mu_B$) or U^{4+} ($3.58\mu_B$) ions. The low value of the observed μ_{eff} may indicate an itinerant electron character of the compound. Obviously, the Kondo and crystalline electric field (CEF) effects may also partly increase the magnitude of the negative Θ_p-value and may reduce the μ_{eff}-value. The presence of the Kondo effect may be presumed basing on the temperature dependence of the electrical resistivity (see below), while the influence of the CEF effect on the behaviour of the susceptibility is not clear, since any information on the CEF levels is not yet available.

The isothermal magnetization measured at temperatures below 3 K (figure 5(a)) is characterized by a break of slope of the $M(H)$ curve at low fields. For larger fields the magnetization follows a monotonic increase but it does not saturate up to 5.5 T, reaching $0.13\mu_B$. Such a field dependence of the magnetization is typical for ferromagnetic clusters. The absence of long-range magnetic ordering at low temperatures is further supported by the analysis of the Arrott plots (figure 5(b)). In simple mean-field ferromagnets, the M^2 versus H/M dependence at various temperatures around T_C should show a series of parallel lines in accordance with the magnetic equation of state of the form $M^2 = A + BH/M$. In the ordered state the coefficient $A > 0$, but $A < 0$ in the paramagnetic state and $A = 0$ at the Curie temperature. For $URh_{0.7}Ru_{0.3}Ge$ the straight lines of $M^2 = A + BH/M$ curves do not give

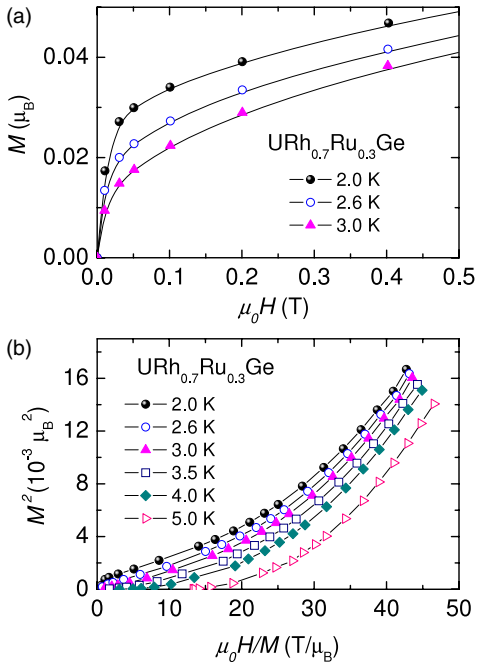

Figure 5. (a) Low-field magnetization curves at 2, 2.6 and 3 K and (b) the Arrott plot for $URh_{0.7}Ru_{0.3}Ge$.

any positive parameter A, being consistent with the lack of a spontaneous magnetization. Thus this rules out long-range ferromagnetic order in this alloy.

The specific heat data are shown in figure 6 in the form C_p/T versus temperature. Below 6 K the C_p/T ratio exhibits an upturn and neither evidences long-range magnetic ordering nor spin-glass freezing. Instead, the specific heat of $URh_{0.7}Ru_{0.3}Ge$ obeys the relationship $C_p = \gamma T + \beta_{ph}T^3 + \delta T^3 \ln(T/T_{sf})$ in the temperature range 1.8–4.1 K. The presence of the last term suggests the existence of spin fluctuations [30]. In this equation, γ is the electronic specific heat coefficient, β_{ph} the phonon coefficient, T_{sf} a characteristic spin-fluctuation temperature, and δ is a constant relating to the coefficient β_{sf} to the $T^3 \ln(T)$ term via $\beta_{sf} = -\delta \ln(T_{sf})$. The fit of the expression $C_p/T = \gamma + \beta T^2 + \delta T^2 \ln(T)$ to the data is shown by the solid line in figure 6(a). From the fit we find $\gamma = 259(2)$ mJ mol^{-1} K^{-2}, $\beta = \beta_{ph} + \beta_{sf} = -9.27(8)$ mJ mol^{-1} K^{-4} and $\delta = 2.95(7)$ mJ mol^{-1} K^{-4}.

The specific heat data collected at temperatures up to 100 K (figure 6(b)) allow us to access the information about the phonon spectrum. We have tried to fit the data using the Debye function. However, a satisfactory fit is achieved only in a limited temperature range 80–100 K (dashed line in figure 6(b)). If we assume that each kind of atom involved has a distinct phonon spectrum we may consider the phonon specific heat to be the sum of at least three Debye spectra, corresponding to those of U, Rh/Ru and Ge atoms. If this assumption is valid for $URh_{0.7}Ru_{0.3}Ge$ we can fit the data in a more extended range, 30–100 K. The result of such a fit with the Debye temperature $\Theta_D^U = 130$ K, $\Theta_D^{Rh/Ru} = 240$ and $\Theta_D^{Ge} = 350$ K, is shown as the solid line in figure 6(b). The average Debye temperature of 240 K corresponds to $\beta_{ph} = 0.422$ mJ mol^{-1} K^{-4}. With the latter value and the coefficients β and δ we can evaluate the spin-fluctuation temperature, $T_{sf} = 27(1)$ K. It is worthwhile noting that the fitting parameters are in a class with those of classical spin fluctuator UAl_2, ($\gamma = 145$ mJ mol^{-1} K^{-2},

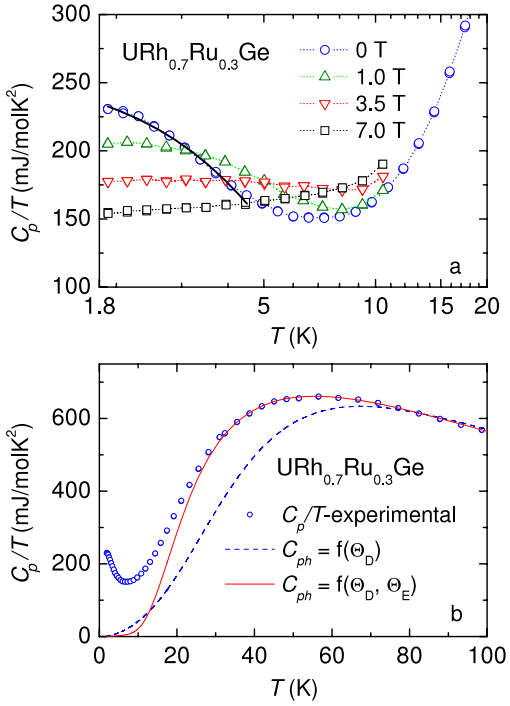

Figure 6. (a) Temperature dependence of the specific heat divided by temperature for $URh_{0.7}Ru_{0.3}Ge$ measured at several magnetic fields. The solid line is the fit. (b) The dashed and solid lines mark the phonon contributions to the high-temperature specific heat data.

$\beta = -3.8$ mJ mol^{-1} K^{-4}, $\delta = 1.67$ mJ mol^{-1} K^{-4} and $T_{sf} = 10.5$ K [29]). We may add that the spin fluctuations remain in Y-doped alloys $Y_{1-x}U_xAl_2$ as long as the non-Fermi-liquid state does not yet form in the composition with $x = 0.875$ [29].

In the framework of the spin-fluctuation theory [30], the upturn in the C_p/T ratio at low temperatures is due to the formation of a narrow peak in the density of states (DOS). Obviously, application of sufficiently high magnetic fields will quench the spin fluctuations, and therefore broaden the DOS peak, resulting in the lowering of C_p/T values. Such a situation seems to be realized in the case of $URh_{0.7}Ru_{0.3}Ge$. At a field of 7 T, C_p/T at 2 K amounts to 153 mJ mol^{-1} K^{-2} compared to the zero-field value of 232 mJ mol^{-1} K^{-2}. Without the effect of spin fluctuations at high enough magnetic fields and neglecting the contribution of the electron–phonon interaction, the ratio C_p/T at 2 K may approximately correspond to γ_0. Using equation (3) we have estimated the value of $N_0(\varepsilon_F)$ to amount to 65 eV^{-1} atom^{-1}. Taking values of $\chi(0)$ and $N_0(\varepsilon_F)$, we can determinate the Stoner enhancement factor S from equation (2): it is about 490. This value is reasonable if one considers the spin-fluctuation theory for the magnetocalorimetric effect [31]. Béal-Monod *et al* [31] predicted the change in specific heat in an applied magnetic field B as

$$\frac{\gamma(B) - \gamma(B=0)}{\gamma(B=0)} = -0.1\frac{S}{\ln S}\left(\frac{\mu_B B}{k_B T_{sf}}\right)^2. \tag{4}$$

Using equation (4), $S = 490$ and $T_{sf} = 27$ K, we have evaluated the theoretical quench rate at a field of 7 T to be about 24.5%, which is consistent with the experimental value of 34.1%.

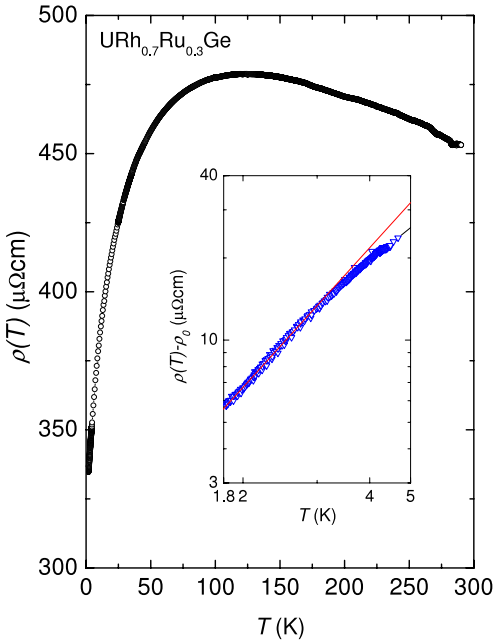

Figure 7. Temperature dependence of the electrical resistivity for $URh_{0.7}Ru_{0.3}Ge$. The inset shows the low-temperature data. The solid line is the fit.

We show in figure 7 the temperature dependence of the electrical resistivity $\rho(T)$. The data are characterized by a maximum at a temperature around 125 K. This behaviour may indicate the presence of the Kondo effect. The presence of the Kondo effect not only results additionally in the negative value of the paramagnetic Curie temperature but may be the reason for the large residual resistivity ρ_0 of the alloy. Obviously, one cannot forget the atomic disorder, which usually increases the ρ_0 value too. It is worthwhile noting that $\rho(T)$ does not obey the T^2 law in the low-temperature range, but rather follows a power law with $T^{1.7}$ (see the inset of figure 7). The observed exponent is close to the value of $3/2$ predicted by Moriya and co-workers [32]. In terms of the self-consistent renormalization model the authors considered the coupling of conduction electrons to antiferromagnetic spin fluctuations, and have provided the relation $\rho \propto T^{3/2}$ for the case of three-dimensional systems at the antiferromagnetic instability. For $URh_{0.7}Ru_{0.3}Ge$, in addition to the contribution of spin fluctuations, short-range order effects, which are evidenced from magnetic data, have to be taken into consideration. A $T^{3/2}$ term in $\rho(T)$ is one of the characteristic features of systems showing short-range magnetic order [33].

4. Concluding remarks

We have studied the magnetic behaviour of $URh_{0.7}Ru_{0.3}Ge$. This alloy exhibits a broad maximum in the ac susceptibility at low temperatures. However, no evidence for either long-range magnetic order or spin-glass freezing was found in the dc magnetization, the specific heat or the electrical resistivity data. From the frequency dependence of the ac susceptibility, the field dependences of the dc magnetization and of the specific heat as well as from the temperature dependence of the resistivity, we infer that short-range magnetic ordering with spin fluctuations can reasonably well describe the magnetic behaviour of the investigated compound.

The investigated alloy is characterized by a relatively low spin-fluctuation temperature and a high Stoner enhancement factor. These physical quantities together with a large ratio C_p/T at low temperatures and the enormous effect of magnetic fields on the specific heat are indicative of strong spin fluctuations. Thus the observed behaviour in $URh_{0.7}Ru_{0.3}Ge$, as a boundary to the NFL state of $URh_{0.62}Ru_{0.38}Ge$, is distinct from that found in $Y_{1-x}U_xAl_2$ [9], $UCu_{5-x}Pd_x$ [23], and $Th_{1-x}U_xPd_2Al_3$ [10], where the spin-glass phase is the inseparable element of the NFL state. In our opinion, the experimental data may provide important information for theoreticians working on models accounting for the NFL behaviour that appears in systems in which the key ingredient leading to the NFL state is a sort of spin fluctuation.

Acknowledgments

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