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Spin-glass behaviour with extended short-range ferromagnetic order in U_2RhSi_3

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Abstract. For U_2RhSi_3, which crystallizes in a disordered derivative of the hexagonal AlB_2-type structure, we present a detailed investigation of magnetic properties by measurements of AC and DC susceptibility, magnetization, magnetic relaxation, specific heat and electrical resistivity. The results provide evidence for the formation of extended short-range ferromagnetic order at the temperature $T_C \approx 25$ K, as indicated by a specific heat anomaly, and for a low-temperature spin-glass state with a freezing temperature $T_f = 19.6$ K. According to systematic specific heat experiments on isostructural spin-glass systems, short-range ferromagnetic correlations above the spin-freezing temperature are observed in U_2RhSi_3 ($T_C \approx 25$ K, $T_f = 19.6$ K) and U_2CoSi_3 ($T_C \approx 10$ K, $T_f \approx 8$ K), but not in U_2PdSi_3 ($T_f = 13.5$ K) and U_2NiSi_3 ($T_f \approx 22$ K). Different degrees of structural disorder of transition metal and silicon atoms, both on the same crystallographic site, seem to be the origin for the distinct different magnetic behaviour.

1. Introduction

Members of the ternary uranium intermetallics U_2TSi_3 (T = Fe, Co, Ni, Ru, Rh, Pd) crystallize in a disordered derivative of the hexagonal AlB_2-type structure (space group $P6/mmm$, No 191, with the sites 1a occupied by Al and 2d by B atoms). T and Si atoms (both on the boron site) are distributed into the trigonal prisms of a primitive hexagonal array of uranium atoms (aluminium site). X-ray and electron diffraction experiments [1, 2] revealed that structural ordering between T and Si atoms occurs and is favoured when the transition metal element has fewer d electrons: it is found to be perfect for U_2RuSi_3 (Ru, $4d^7$, hexagonal superstructure [1]) and decreases gradually from U_2RhSi_3 (Rh, $4d^8$, orthorhombic superstructure [2]) to U_2PdSi_3 (Pd, $4d^{10}$, disordered with a weak tendency to an orthorhombic superstructure [2]). The crystal structures of these three compounds are compared in figure 1. For U_2TSi_3 (T = Fe, Co, Ni) experimental evidence for a crystallographic superstructure has not been reported so far.

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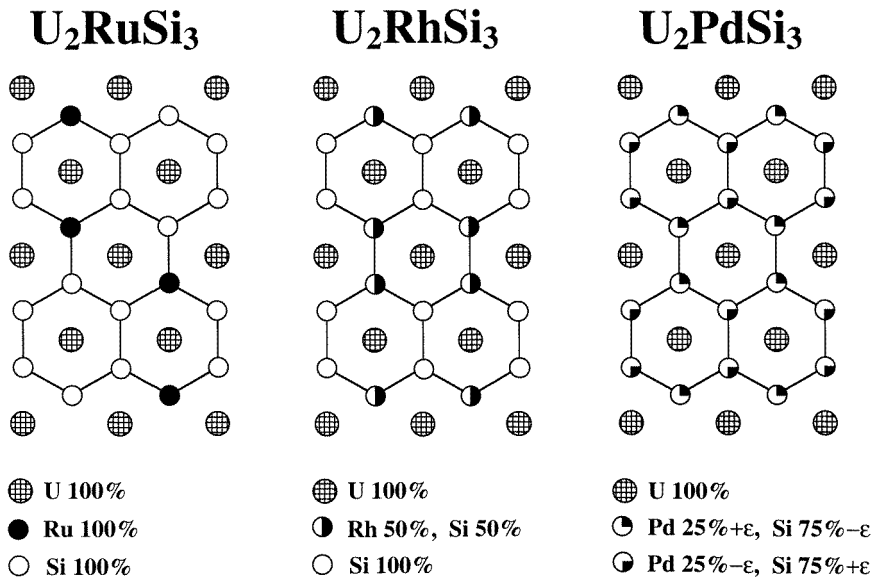

Figure 1. The crystal structures of U_2TSi_3 (T = Ru, Rh, Pd) projected onto the hexagonal *ab*-plane [1, 2]. The height z/c is 0 for U and $\approx 1/2$ for all other atoms. For U_2PdSi_3 the weak tendency to an orthorhombic superstructure of Pd and Si atoms is indicated by $\varepsilon \geqslant 0$.

In U_2TSi_3 systems the introduction of a random distribution of transition metal T and Si atoms on the same crystallographic site can generate frustration via a competition between ferromagnetic and antiferromagnetic U–U exchange interactions. In a ferromagnet or antiferromagnet with weak random frustration the magnetic ordering temperature T_C or T_N decreases with increasing disorder up to a certain level, where the random bond frustration cannot accommodate a percolating ferromagnetic or antiferromagnetic ground state. Such systems often exhibit a spin-glass transition, where the spins freeze cooperatively in a nontrivial pattern that is random in space. Of particular interest are random magnets close to the point in the disorder–temperature phase diagram where the paramagnetic, the ferromagnetic or antiferromagnetic and the spin-glass phase meet and where so-called *reentrant* spin-glass behaviour is observed (e.g., long-range ferromagnetic or antiferromagnetic order between a high-temperature paramagnetic and a low-temperature spin-glass region) [3, 4].

Magnetic properties of U_2TSi_3 (T = Fe, Co, Ni, Ru, Rh, Pd) compounds have been partly investigated [1, 2, 5] by means of magnetization and AC and DC susceptibility measurements. In principle the magnetism in these systems may originate from both the uranium and the transition metal sublattice. However, previous studies [2, 5, 6] suggest that only the uranium atoms, which are completely ordered on one crystallographic site, carry magnetic moments. The compounds U_2RuSi_3 [1, 2] (with a perfect structural order between Ru and Si atoms, see figure 1) and U_2FeSi_3 [5] remain paramagnetic down to 4.2 K and exhibit spin fluctuation behaviour with strong antiferromagnetic U–U exchange interactions, as indicated by a large negative value for the paramagnetic Curie temperature Θ_p obtained from a fit of the Curie–Weiss law to the magnetic susceptibility. In the other four systems U_2TSi_3 (T = Co, Ni, Rh, Pd) the presence of a random distribution of transition metal T and Si atoms on the same crystallographic site varies the electronic environment around the U atoms and introduces random ferromagnetic and antiferromagnetic U–U exchange interactions, which are mediated

by the hybridization between 5f(U) and d(T) electronic states. Such a random frustration gives rise to the formation of the spin-glass state at low temperature in all four systems, while allowing for predominantly ferromagnetic interactions, as indicated by a positive value for the paramagnetic Curie temperature Θ_p [2, 5, 7]. For the UT_2Si_2 silicides, Endstra *et al* [8] have demonstrated that magnetic properties are governed by the strength of the 5f(U)–d(T) hybridization and they proposed an 'f–d hybridization model', based on the determination of the 5f(U)–d(T) overlap. Physical properties of the U_2TSi_3 series might be explained with a similar model.

Obviously, magnetic properties of U_2TSi_3 compounds are strongly influenced by the nature of the transition metal element T, which governs the strength of the hybridization between 5f(U) and d(T) electronic states and consequently the nature of the uranium magnetic state. Therefore, it is essential to collect information about the anomalous magnetic properties of a great number of U_2TSi_3 compounds with AlB_2-type crystal structure and different degrees of atomic disorder. Based on first susceptibility and magnetization data alone the compounds U_2FeSi_3 [5], U_2CoSi_3 [5] and U_2RhSi_3 [2] have all been suggested to be reentrant spin-glass systems. Recently, a more detailed investigation of magnetic properties of U_2PdSi_3 [7] by extensive measurements of DC susceptibility, magnetization, magnetic relaxation, specific heat and electrical resistivity provided clear evidence that U_2PdSi_3 is a simple spin-glass system with a freezing temperature $T_f = 13.5$ K and no ferromagnetic ordering. In this paper we extend the detailed investigation of magnetic properties to U_2RhSi_3, a compound where the degree of structural disorder is smaller than in the previously studied system U_2PdSi_3. We present AC and DC susceptibility, magnetization, magnetic relaxation, specific heat and electrical resistivity data on U_2RhSi_3 and, in order to complete experimental information, specific heat data on U_2CoSi_3 and U_2NiSi_3. Magnetic properties of the spin-glass systems U_2TSi_3 (T = Co, Ni, Rh, Pd) are compared.

2. Experimental details

A polycrystalline sample of U_2RhSi_3 was synthesized by reacting the constituent elements (U: 3N, Pd: 4N, Si: 6N) in an argon arc-furnace and subsequent annealing in an evacuated quartz tube for 72 hours at 800 °C. Within the hexagonal AlB_2-type crystal structure, the room-temperature lattice parameters $a = 4.074$ Å and $c = 3.881$ Å were determined from the x-ray diffraction pattern of the annealed U_2RhSi_3 sample. Measurements of AC susceptibility (2 K $\leqslant T \leqslant$ 30 K, 0.01 Hz $\leqslant \nu \leqslant$ 1000 Hz), DC susceptibility (2 K $\leqslant T \leqslant$ 40 K; -1 T $\leqslant H \leqslant$ 1 T), magnetization (2 K; -1 T $\leqslant H \leqslant$ 1 T) and magnetic relaxation (2 K $\leqslant T \leqslant$ 15 K, $t \leqslant$ 3 h) on U_2RhSi_3 were performed by using a Quantum Design SQUID magnetometer. High-field magnetization experiments up to 23 T on U_2RhSi_3 at 4.2 K were carried out at the High Field Laboratory for Superconducting Materials, Institute for Material Research, Tohoku University in a steady magnetic field. The specific heat of U_2RhSi_3 was measured between 1.6 and 40 K by an adiabatic heat pulse method, and the electrical resistivity between 0.5 and 300 K by the conventional four-terminal DC method. For U_2CoSi_3 and U_2NiSi_3, sample preparation and specific heat experiments were done similarly to the U_2RhSi_3 compound.

3. Results

Figure 2 displays the temperature variation of the magnetic AC susceptibility of U_2RhSi_3 between 15 and 28 K. Both the real (χ') and the imaginary (χ'') components exhibit pronounced

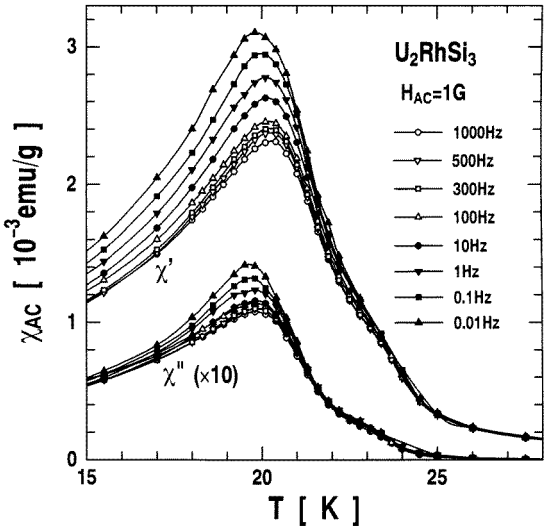

Figure 2. Temperature variation of the AC susceptibility (χ', χ'') for U_2RhSi_3 studied in a magnetic field $H_{AC} = 1$ G at various frequencies ranging from 0.01 to 1000 Hz.

maxima with amplitudes and positions depending on the frequency of the applied magnetic field, especially in the low frequency range. The two susceptibility maxima appear at slightly different temperatures: 19.8 K for χ' and 19.6 K for χ'' (at $\nu = 0.01$ Hz). These results indicate the formation of a spin-glass state in U_2RhSi_3 with the spin freezing temperature $T_f = 19.6$ K (at $\nu = 0.01$ Hz) defined by the peak position of the χ'' anomaly [9]. However, similar to U_2CoSi_3 [5], U_2RhSi_3 is not a simple spin glass, because χ'' does not vanish just above T_f, but remains non-zero up to $T_C \approx 25$ K.

The temperature dependence of the zero-field cooled DC susceptibility ($\chi \equiv M_{ZFC}/H$) of U_2RhSi_3 is shown in figure 3 for magnetic fields between 100 and 5000 G. Similar to U_2PdSi_3 (see figure 1 in [7]) the spin-glass state in U_2RhSi_3 can be observed by a peak in the susceptibility curve at a strongly field dependent temperature $T_f(H)$. With increasing field the peak loses intensity, broadens and the position shifts to lower temperature ending up in a rounded maximum at around 3.2 K ($H = 5000$ G). Above $T_C \approx 25$ K the susceptibility curves of U_2RhSi_3 appear field independent for $H \leqslant 2500$ G with absolute values similar to the data for U_2PdSi_3. Below T_C the M_{ZFC}/H curves of U_2RhSi_3 exhibit a strong field dependence and the amplitudes of the anomalies at T_f are much larger than in U_2PdSi_3 (a factor of 14 at $H = 100$ G). Thus, the formation of ferromagnetic correlations is observed in U_2RhSi_3 at $T_C \approx 25$ K, but not in U_2PdSi_3.

Figure 4 compares the temperature variation of the DC susceptibility ($\chi \equiv M/H$) of U_2RhSi_3 measured in the field cooling (FC) mode and in the zero-field cooling (ZFC) mode for magnetic fields between 2 and 1000 G. The arrows in figure 4 indicate a strongly field dependent characteristic temperature $T_{ir}(H)$, below which a difference is observed between FC and ZFC branches. The M_{FC}/H curves are stable (independent of the time of the measurement), reversible (when heating and cooling in a constant magnetic field) and have a tendency to approach a constant value at low temperatures. In contrast, below T_{ir} the M_{ZFC}/H curves exhibit a metastable and irreversible behaviour (dependence on the elapsed time and the cooling and heating history in the magnetic field). With increasing magnetic field the observed shift to lower temperature is strong for the spin freezing temperature T_f (indicated by the maximum

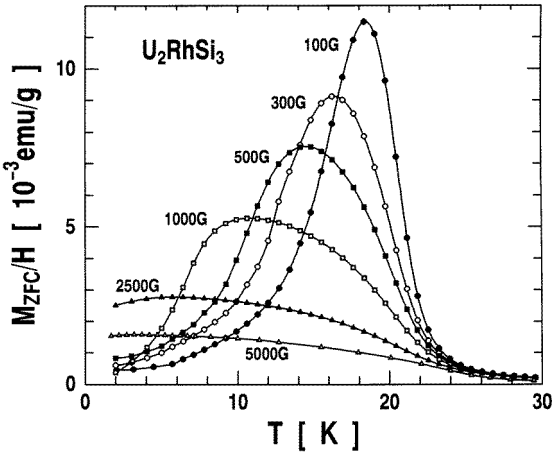

Figure 3. Temperature dependence of the zero-field cooled DC susceptibility ($\chi \equiv M_{ZFC}/H$) of U_2RhSi_3 for magnetic fields between 100 and 5000 G.

of the M_{ZFC}/H curve) and weak for the temperature T_C (indicated by the increase of the M_{FC}/H curve). The temperature T_{ir} drifts from T_C at low magnetic field ($T_{ir} \approx T_C$ at $H = 2$ G) towards T_f with increasing magnetic field ($T_f < T_{ir} \leqslant T_C$). Comparing the data measured at $H = 100$ G reveals again different results for U_2PdSi_3 ($T_{ir} = T_f$, see figure 2 of [7]) and U_2RhSi_3 ($T_{ir} > T_f$, see figure 4).

The magnetization $M(H)$ of U_2RhSi_3 at 4.2 K, shown in figure 5, does not saturate up to 23 T, reaching 1.08 μ_B/U atom. An extrapolation from the almost linear field dependence of $M(H)$ between 6 and 23 T yields a zero-field value of 0.57 μ_B/U atom. A characteristic remanence effect and magnetic relaxation on a macroscopic time scale can be observed for all spin glasses, when changing the magnetic field below the spin freezing temperature T_f [11–13]. For U_2RhSi_3 these features are illustrated in the inset of figure 5 and in figure 6. At 5 K the hysteresis loop of the magnetization of U_2RhSi_3 contains a remanent magnetization $\mu_R = 0.33\ \mu_B$/U atom, which is much larger than the value 0.04 μ_B of U_2PdSi_3 [7]. For the measurement of the isothermal remanent magnetization M_{IRM} as a function of time t, the U_2RhSi_3 sample was first zero-field cooled from a temperature much higher than T_f and T_C to the desired temperature. Then a magnetic field of 0.5 T was applied for 5 minutes and switched off at $t = 0$. As shown in figure 6, below T_f the decay of the remanent magnetization of U_2RhSi_3 is remarkably slow (decrease of only 2.2% after 3 hours at 2 K) and the relaxation time decreases with increasing temperature. Solid lines in figure 6 correspond to a fit to the observed time dependence of M_{IRM} of U_2RhSi_3 using the expression

$$M_{IRM}(T, t) = M_0(T) + \alpha(T) \ln t. \tag{1}$$

The results for the two temperature dependent fitting parameters $M_0(T)$ and $\alpha(T)$ are given in table 1. Compared to U_2RhSi_3, in U_2PdSi_3 the time decay of M_{IRM} is faster (decrease of 5.8% after 3 hours at 2 K, see figure 3 of [7]) and an additional exponential term had to be added to the expression of equation (1) to obtain a good fit to the M_{IRM} curve at short time ($t < 400$ s).

The specific heat data of U_2RhSi_3 are displayed in figure 7. The absence of long-range spatial magnetic ordering at the spin-glass freezing temperature $T_f = 19.6$ K is confirmed by the absence of specific heat anomaly. However, at the temperature $T_C \approx 25$ K a specific heat anomaly is observed, which contains a very small amount of magnetic entropy of

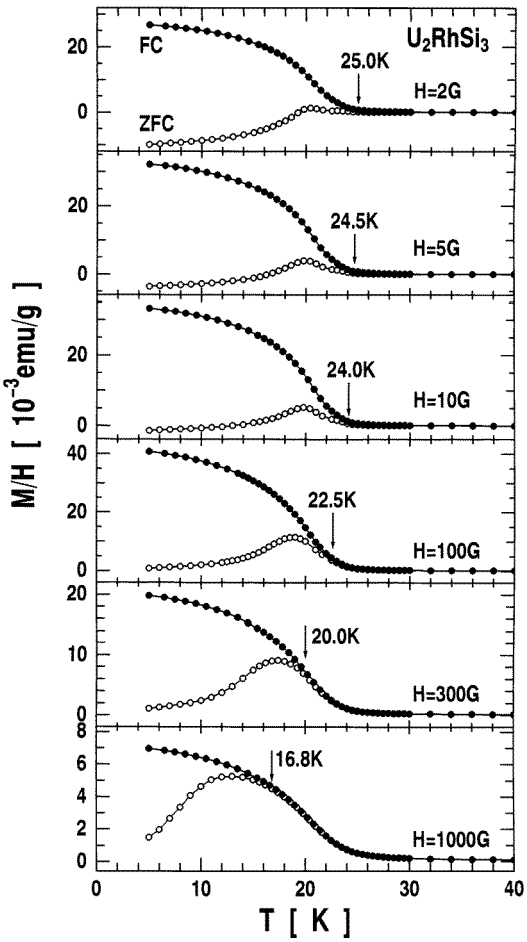

Figure 4. Temperature variation of the DC susceptibility ($\chi \equiv M/H$) of U_2RhSi_3 measured in the field cooling (FC,●) and in the zero-field cooling (ZFC,○) mode for magnetic fields between 2 and 1000 G. The arrows indicate the temperature T_{ir}, below which a difference is observed between the FC and ZFC branches. Note that the observed negative value of M_{ZFC}/H at low fields and low temperatures arises because the sample is cooled in a net negative magnetic field, present above the superconducting solenoid, as explained in detail in [10].

Table 1. Results of a fit using equation (1) to the time dependence of the isothermal remanent magnetization $M_{IRM}(t)$ of U_2RhSi_3 shown in figure 6.

T [K]	M_0 [emu g^{-1}]	α [emu g^{-1}]
2	6.266	0.0252
5	5.639	0.0254
10	4.315	0.0415
15	2.191	0.0462

$S_{mag}/R = 0.0132$ per U atom (corresponding to only 1.9% of the value ln 2 expected for magnetic ordering of a crystalline-electric-field ground-state doublet). In contrast to U_2RhSi_3, no specific heat anomaly is found in U_2PdSi_3 (see figure 6 in [7]).

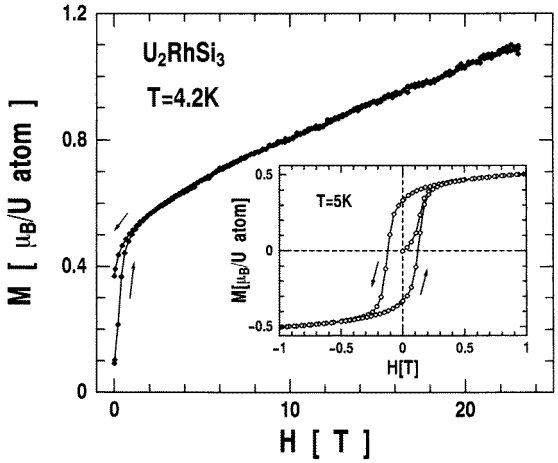

Figure 5. High-field magnetization $M(H)$ up to $H = 23$ T of U_2RhSi_3 at 4.2 K. The hysteresis loop at 5 K is shown in the inset.

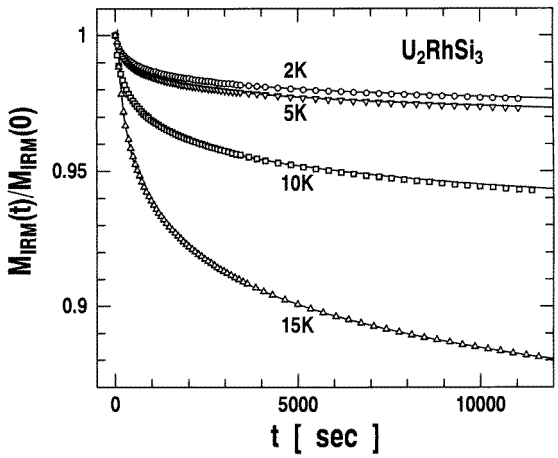

Figure 6. Time dependence of the isothermal remanent magnetization $M_{IRM}(t)/M_{IRM}(0)$ of U_2RhSi_3 measured at different temperatures below T_f. The solid lines represent least-squares fits using equation (1).

The temperature dependence of the electrical resistivity $\rho(T)$ is found to be very weak for both compounds U_2RhSi_3 (see figure 8) and U_2PdSi_3 (see figure 6 in [7]), with no $\rho(T)$ anomalies at T_f or T_C. Due to scattering at structural disorder, which is present in both compounds and higher in U_2PdSi_3, the electric resistivity is dominated by a large temperature independent contribution of the order of 147 $\mu\Omega$ cm for U_2RhSi_3 and 255 $\mu\Omega$ cm for U_2PdSi_3. The resulting small values for the residual resistivity ratio $\rho(T = 300\ \text{K})/\rho(T = 0.5\ \text{K})$ are 1.19 for U_2RhSi_3 and 1.04 for U_2PdSi_3. Interestingly, a minimum of $\rho(T)$ has been observed in U_2RhSi_3 (at 6 K, below T_f) and U_2PdSi_3 (at 26 K, above T_f), but not in U_2CoSi_3 down to 1.6 K [14].

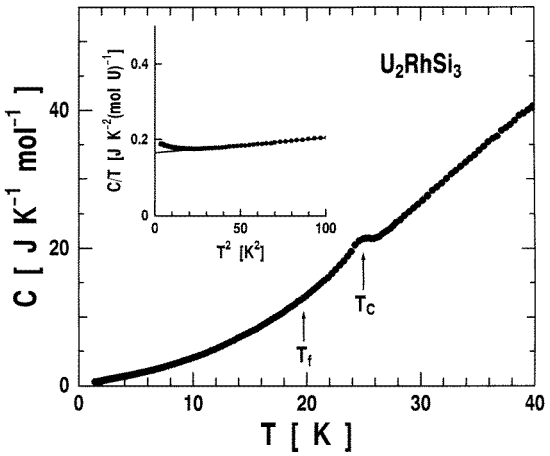

Figure 7. Temperature variation of specific heat C_p of U_2RhSi_3. The inset shows the C/T versus T^2 plot.

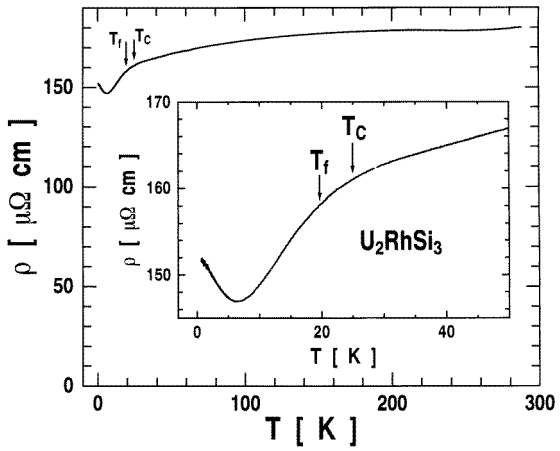

Figure 8. Temperature dependence of the electrical resistivity $\rho(T)$ of U_2RhSi_3 between 0.5 and 300 K. The inset displays the $\rho(T)$ data at low temperatures in an expanded scale.

4. Discussion

Crystallographic and magnetic properties of the compounds U_2TSi_3 (T = Fe, Co, Ni, Ru, Rh, Pd), with disordered hexagonal AlB_2-type crystal structure, are compared in table 2. The detailed experimental results presented on U_2PdSi_3 (in a previous study [7]) and on U_2RhSi_3 (in this work) reveal distinct differences in the magnetic properties of the two compounds, which reflect the different degree of structural disorder of non-magnetic atoms (see figure 1) and the different strength of random frustration of U–U interactions. U_2PdSi_3, with completely ordered U atoms on the Al-site and a random distribution of Pd and Si atoms on the B site, is found to be a simple spin-glass system with the freezing temperature $T_f = 13.5$ K and no detectable ferromagnetic ordering. In contrast, U_2RhSi_3, with completely ordered U atoms on the Al site and an orthorhombic superstructure of the Rh and Si atoms on the B site, can be

Table 2. Crystallographic and magnetic properties of U_2TSi_3 (T = Fe, Co, Ni, Ru, Rh, Pd) compounds with disordered hexagonal AlB_2-type crystal structure. a, c: room temperature lattice parameters; μ_{eff}: effective magnetic U moment and Θ_p: paramagnetic Curie temperature, obtained from Curie–Weiss fit to the magnetic susceptibility; μ_R: remanent magnetization at 5 or 4.2 K; γ: linear term of the specific heat for $T \to 0$ K; T_C: specific heat anomaly; T_f: spin-glass freezing temperature.

	U_2FeSi_3 [5]	U_2RuSi_3 [1, 2]	U_2CoSi_3 [5]	U_2RhSi_3 [this work]	U_2NiSi_3 [5]	U_2PdSi_3 [7]
a [Å]	4.004	4.074	3.988	4.074	3.979	4.083
c [Å]	3.864	3.855	3.883	3.881	3.949	3.932
c/a	0.965	0.946	0.974	0.953	0.992	0.963
μ_{eff} [μ_B/U atom]	2.77	3.02	2.03	2.32[a]	2.38	2.39[a]
	3.35[a]		2.01[a]		2.41[a]	
Θ_p [K]	−86	−130	0	17[a]	11	7[a]
	−132[a]		2[a]		32[a]	
μ_R [μ_B/U atom]	0	0	0.05	0.33	0.31	0.04
				0.31[a]		0.03[a]
γ [mJ K^{-2} (mol U)$^{-1}$]	97[c]	115[c]	155[b]	115	158[b]	180
T_C [K]	—[c]	—[c]	10	25	—[b]	—
T_f [K]	—	—	8	19.6	22	13.5
				20.3[a]		10.2[a]

[a] [2].
[b] [this work].
[c] [14].

classified as a spin-glass system ($T_f = 19.6$ K is indicated by a peak in the AC susceptibility, figure 2) with the formation of short-range ferromagnetic order at $T_C \approx 25$ K, as indicated by a peak in the specific heat (figure 7) and by an increase of the magnetic susceptibility (figure 3). The specific heat anomaly contains a magnetic entropy of $S_{mag}/R = 0.019$ ln 2 per U atom, which seems to be much too small for the development of usual long-range ferromagnetic order. But to determine the actual value of the ferromagnetic correlation length in U_2RhSi_3, a future neutron scattering experiment must be awaited. The fact that the low-field FC and ZFC susceptibility is split up to $T_C \approx 25$ K (figure 4) means that the onset of ferromagnetic correlations in U_2RhSi_3 shows glassy features, which are continuously merged and pushed to lower temperature with increasing applied magnetic field.

Our results suggest that, compared to U_2RhSi_3, the system U_2PdSi_3 is located further away from the tricritical point in the disorder–temperature phase diagram, where the paramagnetic, the ferromagnetic and the spin-glass phase meet. The place of U_2RhSi_3 ($T_C \approx 25$ K, $T_f = 19.6$ K) is between a simple spin-glass system (an example is U_2PdSi_3, $T_f = 13.5$ K [7]) and a true reentrant spin-glass system (examples are $Eu_{0.54}Sr_{0.48}S$, $T_C = 5$ K, $T_f = 2$ K [3, 11, 15] and amorphous $(Fe_{1-x}Mn_x)_{75}P_{16}B_6Al_3$, $0.22 \leqslant x \leqslant 0.26$, 200 K $< T_C <$ 300 K, $T_f \approx 20$ K [3, 16, 17]). Magnetism in U_2PdSi_3 is characterized by one temperature (T_f) and in U_2RhSi_3 by two temperatures (T_f and T_C).

For U_2RhSi_3, figure 9 illustrates the pronounced dependence of the freezing temperatures $T_f(\chi')$ and $T_f(\chi'')$ on the frequency ν, based on the peak maximum of the AC susceptibility curves shown in figure 2. The initial frequency shift $\delta T_f(\chi'')$ was calculated as

$$\delta T_f(\chi'') = \Delta T f(\chi'')/[T_f(\chi'')\,\Delta \log \nu] = 0.008 \pm 0.001. \qquad (2)$$

This value for U_2RhSi_3 is comparable to those reported for other metallic spin-glass systems, e.g., 0.005 for CuMn [13], 0.010 for AuFe [13] and 0.025 for URh_2Ge_2 [18]. Figure 10 displays the field dependence of the freezing temperature T_f for U_2RhSi_3 and U_2PdSi_3,

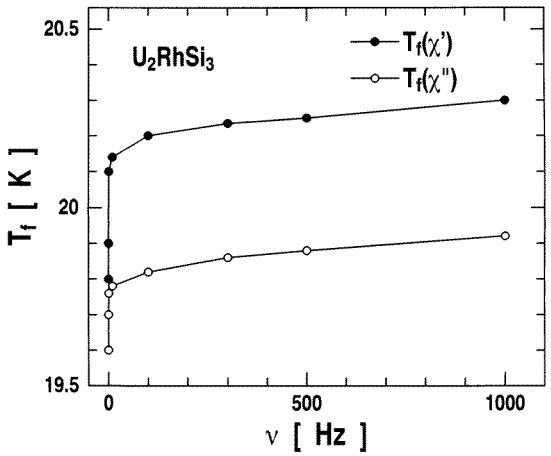

Figure 9. Dependence of the freezing temperatures $T_f(\chi')$ and $T_f(\chi'')$ on the frequency ν for U_2RhSi_3.

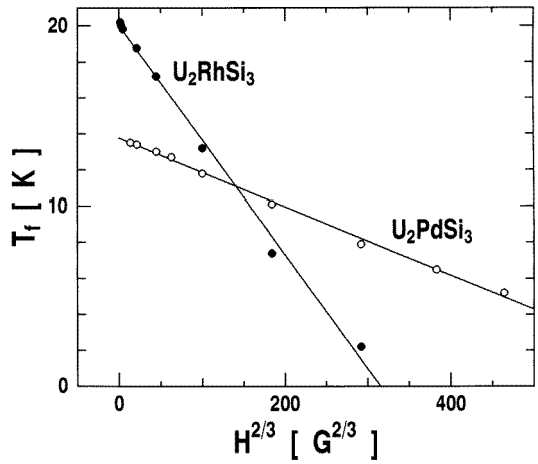

Figure 10. The field dependence of the freezing temperature T_f for U_2RhSi_3 and U_2PdSi_3, plotted as T_f versus $H^{2/3}$.

plotted as T_f versus $H^{2/3}$. Here T_f was determined by the peak maximum of the zero-field cooled susceptibility M_{ZFC}/H curve (data of figures 3, 4 and of figure 1 of [7]). For both compounds, T_f approximately follows an $H^{2/3}$-law, which has also been observed in the spin-glass system UCuSi [19] and has been predicted by the mean-field spin-glass model [20]. The field dependence of T_f is stronger in U_2RhSi_3 than in U_2PdSi_3.

[2] presents nice experimental work on the determination of the crystallographic superstructure of U_2RhSi_3 (see figure 1) and in addition some measurements of magnetic properties of U_2RhSi_3 and U_2PdSi_3. The AC susceptibility data of [2]:

$$U_2RhSi_3,\ \nu = 125\ \text{Hz}: \qquad T_f(\chi') = T_f(\chi'') = 20.3\ \text{K}$$

are not in very good agreement with our results (see figure 9):

$$U_2RhSi_3,\ \nu = 125\ \text{Hz}: \qquad T_f(\chi') = 20.2\ \text{K} > T_f(\chi'') = 19.8\ \text{K}.$$

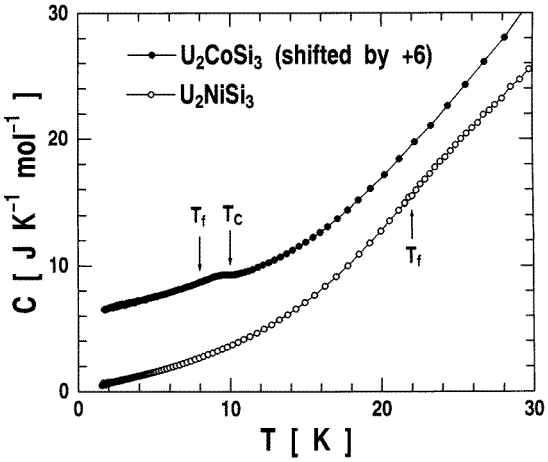

Figure 11. Temperature variation of the specific heat C_p of U_2CoSi_3 and U_2NiSi_3.

In [2] the temperatures $T_C(M_{FC}) = 19.5$ K for U_2RhSi_3 and $T_C(M_{FC}) = 15.5$ K for U_2PdSi_3 were determined from the inflection point of the increase of the field cooled magnetization $M_{FC}(T)$ measured at $H = 1000$ G. For U_2RhSi_3 such a value is consistent with our magnetization data (see figure 4). But information about $T_C(M_{FC})$ is not accurate enough to decide about the existence of long-range or short-range ferromagnetic order above the spin freezing temperature and specific heat or neutron diffraction measurements are necessary.

Specific heat data measured for U_2CoSi_3 and U_2NiSi_3 are shown in figure 11. Also in U_2CoSi_3 ($T_C \approx 10$ K, $T_f \approx 8$ K [5]) short-range ferromagnetic correlations develop above the spin freezing temperature. But compared to U_2RhSi_3, in U_2CoSi_3 the specific heat anomaly near T_C contains a smaller amount of magnetic entropy per U atom of $S_{mag}/R = 0.0132 = 0.011 \ln 2$, indicating that the ferromagnetic correlations are weaker. Our U_2NiSi_3 sample with no specific heat anomaly below 40 K is found to be a simple spin-glass system ($T_f \approx 22$ K [5]) with no detectable ferromagnetic ordering, similar to U_2PdSi_3. Indeed, the AC susceptibility data of [5] reveal that above T_f the decrease of χ'' towards zero is much steeper in U_2NiSi_3 (see figure 7 in [5]) than in U_2CoSi_3 (see figure 5 in [5]). It is known that the disordered hexagonal AlB_2-type crystal structure is adopted within a large composition range $U_2T_{1-x}Si_{3+x}$ (for $x > 0$ and $x < 0$), which is an origin of the sample dependence of magnetic properties. A striking example for such a sample dependence is the result of a neutron diffraction study on a U_2NiSi_3 single crystal [6], which determined long-range ferromagnetic order ($\mu(U) = 0.6(1)\ \mu_B$ oriented perpendicular to the hexagonal c-axis) below 30 K, instead of spin-glass freezing. The magnetic entropy released at such a ferromagnetic phase transition gives rise to a specific heat anomaly, which was not observed for our U_2NiSi_3 sample.

In summary, the spin-glass systems U_2TSi_3 (T = Co, Ni, Rh, Pd) crystallize in a disordered derivative of the hexagonal AlB_2-type structure, with a complete order of U atoms on the Al site and a random distribution of transition metal T and Si atoms on the B site. Extensive experimental information presented on U_2PdSi_3 (in a previous study [7]) and on U_2RhSi_3 (in this work) reveals distinct differences in the magnetic properties of the two compounds: U_2PdSi_3, with a higher degree of structural disorder of Pd and Si atoms, is found to be a simple spin-glass system with a freezing temperature $T_f = 13.5$ K and no detectable magnetic order. U_2RhSi_3, with a lower degree of structural disorder of Rh and Si atoms, can be

classified as a spin-glass system ($T_f = 19.6$ K) with extended short-range ferromagnetic order below $T_C \approx 25$ K. Systematic specific heat experiments reveal that short-range ferromagnetic correlations above the spin-freezing temperature are observed in U_2RhSi_3 and U_2CoSi_3, but not in U_2PdSi_3 and U_2NiSi_3.

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