Investigation of the spin glass transition in a low U doped YRu_2Si_2 sample

2003 J. Phys.: Condens. Matter 15 4613

(http://iopscience.iop.org/0953-8984/15/26/311)

INSTITUTE OF PHYSICS PUBLISHING JOURNAL OF PHYSICS: CONDENSED MATTER

J. Phys.: Condens. Matter **15** (2003) 4613–4621 PII: S0953-8984(03)55416-9

Investigation of the spin glass transition in a low U doped YRu_2Si_2 sample

M Očko[1], **Dj Drobac**[1], **J-G Park**[2,3], **Z Samardžija**[4] **and K Zadro**[5]

[1] Institute of Physics, Bijenička c. 46, PB 304, 10000 Zagreb, Croatia
[2] Department of Physics and Institute of Basic Science, Sung-Kyun-Kwan University, Suwon 440-746, Korea
[3] Centre for Strongly Correlated Materials Research, Seoul National University, Seoul 151-742, Korea
[4] Jožef Stefan Institute, Jamova 39, SI-1000 Ljubljana, Slovenia
[5] Department of Physics, University of Zagreb, Bijenička c. 32, 10000 Zagreb, Croatia

E-mail: ocko@ifs.hr

Received 31 October 2002, in final form 6 May 2003
Published 20 June 2003
Online at stacks.iop.org/JPhysCM/15/4613

Abstract

We have made extensive studies of a dilute U doped YRu_2Si_2 alloy using several experimental techniques such as ac and dc magnetic susceptibility, dc resistivity and microstructural investigations. The real part of the ac susceptibility shows a maximum at around 6.75 K for $f = 7$ Hz. With increasing frequency, the temperature of this maximum increases while, at the same time, the magnitude of the maximum decreases, which is a typical spin glass behaviour. The magnitude of the imaginary part shows frequency-dependent behaviour too. We show that most of the imaginary part of the susceptibility, at least over the range of the frequencies we used, comes from the conduction electrons. Zero-field cooling and field cooling dc magnetization measurements also indicate the presence of a spin glass transition at about the same temperature. We also found that relaxation processes below the freezing temperature show $\ln(t + t_O)$ behaviour. We discuss our findings in light of the origin of the spin glass behaviour and compare them with other similar studies.

1. Introduction

URu_2Si_2 is one of the most intensively studied heavy fermion compounds with a very weak antiferromagnetic transition at 17.5 K with $\mu_{ord} = 0.04\ \mu_B$ and a superconducting transition around 1.2 K [1]. Although the superconducting transition itself is a very interesting subject, the antiferromagnetic transition, in particular its nature, has drawn much more interest. With the relatively moderate γ value ($\gamma = 112$ mJ mol^{-1} K^{-2}), URu_2Si_2 is a strongly correlated electronic system. However, it is surprising that theories based on a localized picture of 5f electrons seem to explain successfully most bulk properties, even at a quantitative level [2]. This localized picture was recently tested against an inelastic neutron scattering experiment and seems to be in reasonably good agreement with the data, at least at a qualitative level [3]. It is important, therefore, to understand well the magnetic properties of the U single ions. Up to now only a few investigations have been made on the alloys containing a small amount of U dissolved in a nonmagnetic matrix like YRu_2Si_2 or $ThRu_2Si_2$ and found anomalous features at low temperature [4–7]. In [4] and [5], these low temperature anomalies were explained in terms of a two-channel Kondo model for both $U_xTh_{1-x}Ru_2Si_2$ and $U_xY_{1-x}Ru_2Si_2$ with $x < 0.08$, i.e. single-ion properties. On the other hand, in [6] a scenario invoking proximity effects near a quantum critical point was proposed as an explanation for the low temperature anomaly in $U_xTh_{1-x}Ru_2Si_2$ for $x = 0.01$. It is also to be noted that, for $x > 0.1$ of $U_xTh_{1-x}Ru_2Si_2$, some spin glass characteristics were reported [7]. An underlying fundamental question common to all these investigations is why does U not produce the usual ordinary, single-channel, Kondo effect, for example a logarithmic upturn in the resistivity in the dilute limit in the matrix like YRu_2Si_2, although such a feature was observed in other dilute U alloys such as $U_xY_{1-x}Pd_3$ ($x < 0.3$) [8]. One can ask if the absence of the single-channel Kondo scattering is due to the interactions which eventually lead to spin glass behaviour [7], or due to the multi-channel scattering [5].

As a part of our experiments on URu_2Si_2, we have studied the alloying effects of URu_2Si_2 by doping both the U and Si sites [9, 10]. Among these experiments, Y doping is particularly interesting because, in spite of its very weak nature, the antiferromagnetic transition is seen to survive up to 70% Y doping [10]. In this paper we present ac, dc magnetic and resistivity measurements together with microstructural investigations on a dilute U doped YRu_2Si_2 sample in order to examine the properties of $U_xY_{1-x}Ru_2Si_2$ at very low U concentrations and, if possible, to shed light on the controversial reports in [4–7].

2. Preparation of the samples and microstructural investigations

Constituent elements were melted together on a water-cooled Cu heath under an Ar atmosphere. Samples, encapsulated in evacuated quartz tubes, were annealed at 600 °C for two days and then at 800 °C for five days. In this paper we show the results on two samples: YRu_2Si_2 and $(U_{0.08}Y_{0.92})Ru_2Si_2$.

Microstructural and elemental compositions of the alloy have been analysed using a scanning electron microscope (SEM, Jeol JSM 5800) and energy dispersive x-ray spectroscopy (EDXS, Oxford–Link ISIS 300). Both SEM and EDXS analysis have shown that, besides the main phase, there exists a small amount of a secondary phase embedded in the matrix. One of the micrographs is shown in figure 1. Quantitative analysis of the EDXS spectra from the matrix phase has shown that it consists of 39.0 ± 0.8 at.% Si, 41.0 ± 0.4 at.% Ru, 19.3 ± 0.2 at.% Y and 0.7 ± 0.05 at.% U. Consequently, the calculated formula of the alloy could be written as $U_{0.035}Y_{0.965}Ru_2Si_2$. The secondary phase is found to consist of 33 at.% Si, 53 at.% Ru and 14 at.% U without Y. The nominal concentration of our sample corresponds to $x = 0.08$ and in the following we used the nominal composition to denote this alloy.

Comparing this value with EDES analysis we estimated that there is about 7% of the secondary phase with respect to the matrix phase. Image analysis of several SEM micrographs, which represent the microstructure of the sample, indicates 7% of the secondary phase in the alloy.

We also note that the Debye–Scherrer method of x-ray diffraction shows that two samples form in a singe phase with little trace of a secondary phase.

Figure 1. A backscattered electron micrograph of the dilute U doped YRu_2Si_2 alloy: grey (matrix phase), white (secondary U-rich phase) and black (pores). (We note that this figure shows a secondary phase more than the average value.)

3. AC susceptibility measurements and analysis of results

AC susceptibility has been measured using a CryoBIND susceptometer at $f = 7, 28.4, 77$ and 231 Hz with the driving field of 14 Oe. The dimensions of a YRu_2Si_2 sample, a nonmagnetic parent compound, were $a = 1.77$ mm, $b = 1.48$ mm and $c = 10.7$ mm while those of a magnetic alloy, $(U_{0.08}Y_{0.92})Ru_2Si_2$, were $a = 1.45$ mm, $b = 1.62$ mm and $c = 8.0$ mm. The measurements were performed with the driving field parallel to the long sample axis. The absolute values of the susceptibility (in SI units) were determined by comparing the ac signals of our sample to the signals of a Nb sample of similar dimensions in a superconducting state ($\chi' = -1$). The obtained values of the susceptibility were small ($\chi' \approx 10^{-3}$ in SI units) and therefore corrections to the demagnetization effect are negligible.

In figure 2, we show the real (χ') and imaginary part (χ'') of the ac susceptibility of the $(U_{0.08}Y_{0.92})Ru_2Si_2$. As one can see, there is a maximum in χ' at 6.75 K for 7 Hz. With increasing the frequency to 231 Hz, the temperature of this maximum increases to 7.15 K and, simultaneously, the magnitude of the maximum decreases while χ' is virtually independent of frequency above 10 K. These observations, consistent with the typical spin glass behaviour [11, 12], indicate strongly the existence of a spin glass state at low temperatures. It is also interesting to note that χ'' shows a step-like behaviour, which is again characteristic of spin glass as pointed out in [11]. However, unlike those similar investigations the magnitude of the imaginary part depends strongly on frequency in our case. In the analysis performed below we explain this experimental observations using Landau–Lifshitz theory [13]. It is worthwhile noting that our subsequent analysis of contributions from U ions to the imaginary part of the ac susceptibility $\Delta\chi''$ shows a peak, which we ascribe to spin glass behaviour in agreement with theoretical predictions [14].

For comparison, we show the ac susceptibility data for YRu_2Si_2 measured in the same condition (see the inset of figure 2). Since YRu_2Si_2 is a nonmagnetic intermetallic compound and a relatively good conductor, one expects that the ac susceptibility follows the predictions of a theory on the propagation of low frequency electromagnetic waves in a conducting medium [13]. According to this theory, ac susceptibility of a conducting medium can be

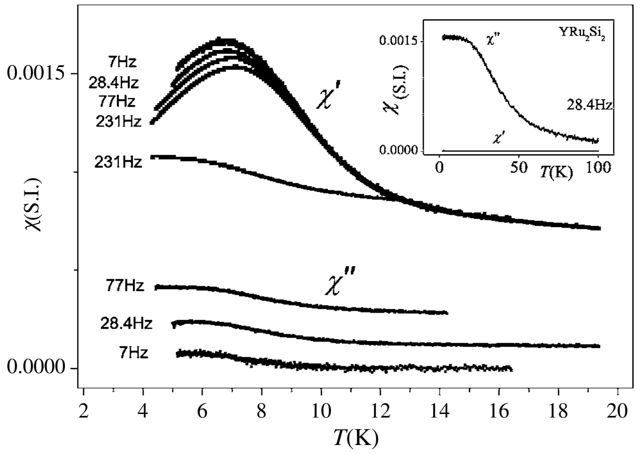

Figure 2. The ac susceptibility of the U dilute sample versus temperature for several driving frequencies. Inset: the ac susceptibility of the nonmagnetic compound YRu_2Si_2 versus temperature for $f = 28.4$ Hz.

expressed in terms of a penetration depth:

$$\delta = (\rho/\pi\mu f)^{1/2}, \tag{1}$$

where ρ is the resistivity, μ is the permeability of medium and f is the frequency. Since the susceptibility of nonmagnetic materials is very small, we can replace μ with μ_0. Substituting the corresponding values into equation (1), one gets

$$\delta \approx 50(\rho/f)^{1/2}\,\text{mm}, \tag{1$'$}$$

where ρ is given in $\mu\Omega$ cm and f in Hz. With the resistivity value of 1.47 $\mu\Omega$ cm for YRu_2Si_2 at the lowest temperature, one has $\delta \approx 11$ mm at 28.4 Hz. For the U doped YRu_2Si_2 alloy, δ is even higher since its resistivity is approximately 20 times larger at 2 K than the resistivity of the pure YRu_2Si_2. Furthermore, δ increases with temperature since ρ increases with temperature. As the cross sections of our samples are 1.77×1.48 mm^2 for the nonmagnetic sample and 1.45×1.62 mm^2 for the doped one, one may assume safely that our experiments were performed in the low shielding limit ($a, b \ll \delta$). In this limit, ac susceptibility of a cylindrical sample with a radius r in a longitudinal field is given by the following formulae:

$$\chi' \approx -(r/\delta)^4/12 \tag{2a}$$

and

$$\chi'' \approx (r/\delta)^2/4. \tag{2b}$$

Equation (2) explains why the measured signals of χ'' for the nonmagnetic sample were small and χ' is almost zero.

If we approximate our samples with a cylindrical shape with the same size of cross section, we get the following result:

$$\chi'' \approx (ab/31\,800)(f/\rho), \tag{2b'}$$

where a and b are in mm, f in Hz, ρ in $\mu\Omega$ cm and χ'' in SI units (dimensionless). Thus χ'' is proportional to frequency and inversely proportional to ρ. Interestingly enough, our data

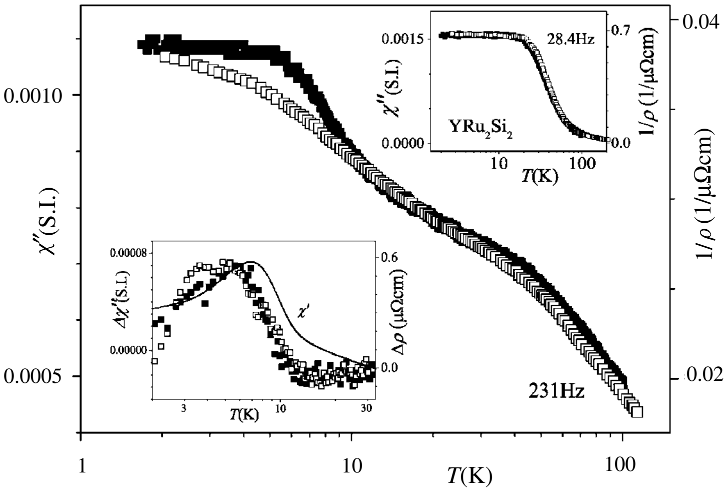

Figure 3. The imaginary part of the ac susceptibility measured at $f = 231$ Hz, χ'' (full squares) and $1/\rho$ (open squares) versus temperature for the magnetic alloy. Upper inset: the same plot for YRu_2Si_2 (the ac susceptibility of YRu_2Si_2 was measured at $f = 28.4$ Hz). Lower inset: the contribution of U ions to the imaginary part of the ac susceptibility, $\Delta\chi''$, calculated using equation (3) (full squares) and an anomalous contribution to the resistivity $\Delta\rho$ (open squares) of the same alloy. The line represents χ' in arbitrary units.

from the nonmagnetic sample show that it is indeed the case. In the upper inset of figure 3, we show that χ'' measured with $f = 28.4$ Hz follows the temperature dependence of the inverse dc resistivity over a wide temperature range (2–100 K). The value of $[\chi''/(1/\rho)]/f$ taken from the experimental results is 8.3×10^{-5} $\mu\Omega$ cm s at 100 K while the calculated value using equation ($2b'$) is 8.2×10^{-5} $\mu\Omega$ cm s, in very good agreement with the experimental results. Equation ($2b'$) also predicts that χ'' is proportional to f. If one divides χ'' for the magnetic alloy at 12 K by frequency, one gets values 0.54×10^{-6}, 4.0×10^{-6}, 3.8×10^{-6} and 3.7×10^{-6} s for $f = 7$, 28.4, 77 and 231 Hz, respectively. Thus χ'' scales roughly with frequency at the higher temperatures, except for the lowest frequency.

The proportionality between χ'' and $1/\rho$ is also found to hold down to 10 K for the magnetic alloy. In figure 3 we plot χ'' measured at $f = 231$ Hz. At this frequency, we have a better signal-to-noise ratio in our system and, more importantly, the ratio b/δ is approximately the same as it is for the nonmagnetic YRu_2Si_2 sample at $f = 28.4$ Hz. The experimental value of $[\chi''/(1/\rho)]/f$ at 100 K for the U doped sample is 5.9×10^{-5} $\mu\Omega$ cm s while the theoretical value from equation ($2b'$) is 14.4×10^{-5} $\mu\Omega$ cm s. We think that this difference between the theoretical and experimental values comes mainly from a large uncertainty in determination of the absolute value of the susceptibility and the resistivity. For example, our resistivity measurement by the four-point method has about 10% of errors due to inaccurate determination of the dimensions of the samples. However, a much larger uncertainty in fact comes from the large porosity inside the sample, as seen in our SEM studies. In spite of these problems in determining the absolute values of ρ and χ'', the Landau–Lifshitz theory is certainly able to reproduce a linear relationship between $1/\rho$ and χ'' over the whole temperature range measured, as one can see clearly in the upper inset of figure 3. This fact is important in our further analysis of the results for the magnetic alloy.

As one can see in figure 3, there is also some disagreement between χ'' and $1/\rho$ below 10 K for the magnetic sample. In order to analyse this deviation, we assume that there are two contributions to the imaginary part of the susceptibility:

$$\chi'' = \chi''_e + \Delta\chi''. \tag{3}$$

χ''_e, representing the conduction electron contribution to χ'', is given by the following equation:

$$\chi''_e = k(1/\rho). \tag{3a}$$

The constant $k = \chi''/(1/\rho)$ can be calculated at higher temperatures where equation (3*a*) holds reasonably well. We think that $\Delta\chi''$ represents the magnetic contributions of U ions to χ''. Unlike χ'', $\Delta\chi''$ shows a peak at lower temperature (5.8 K) than the peak in χ' (see the lower inset of figure 3). Both facts are in agreement with theoretical calculations [14]. What is also interesting is that there is an anomalous behaviour in the resistivity [10]. In the lower inset of figure 3, we plot the anomalous contribution to the resistivity using the data published in [9], $\Delta\rho$ (open squares). The peak in $\Delta\rho$ is located at slightly lower temperature than the peak in $\Delta\chi''$, which we believe is partly due to the fact that $\Delta\rho$ is measured by a dc technique.

In order to analyse further the frequency dependence of the ac susceptibility, we have used the Vogel–Fulcher law:

$$\tau = \tau_o \exp[T_A/(T_f - T_0)], \tag{4}$$

where τ_o is the characteristic relaxation time and $T_0(T_f)$ is the peak position of χ' at $f = 0$ (nonzero f). $E_A = k_B T_A$ is the characteristic energy scale of the activation barrier for a relaxation process. The results of this analysis with $\tau_o = 10^{-13}$ s are shown in figure 4. The parameters we obtained from the analysis: $T_0 = 4.4$ K and $T_A = 66$ K put our magnetic alloy closer to amorphous $AlGd_{10\%}$, $T_0 = 3.8$ K and $T_A = 74$ K (a RKKY spin glass) than to amorphous $AlGd_{25\%}$, $T_0 = 10.58$ K and $T_A = 27.5$ K (a frustrated spin glass) [15]. Since T_0 is very close to T_f, we can conclude that the effect of the RKKY interaction is relatively strong in our magnetic sample. One can also compare the amplitudes of the RKKY interactions, V_0, by using the relation of $E_A \sim V_0 x'$, where x' is the fraction of magnetic ions [15]. The ratios E_A/x' are 110, 740 and 1885 for $AlGd_{25\%}$, $AlGd_{10\%}$ and our sample (we used $x' = 0.035/5$ for our sample), respectively. The ratio E_A/x' for a canonical RKKY spin glass, $CuMn_{3.3\%}$, is 1200. We have also calculated a relative change in the freezing temperature per frequency: $\Delta T_f/(T_f \Delta \log f) = 0.038$. This value is about ten times larger than that obtained for $Cu_{1-x}Mn_x$. In spite of this last disagreement we may conclude that a strong RKKY interaction leads to the observed transition of spin glass nature in our magnetic sample. Here we can add that some quasicrystals have been reported to have spin glass transitions with similarly large frequency dependence, as observed in our sample [16].

4. DC magnetic measurements

DC magnetic measurements were performed on a SQUID magnetometer (Quantum Design's MPMS5). The temperature dependence of the magnetic moment was measured for both zero-field cooling (ZFC) and field cooling (FC) with a field of 100 Oe. We have repeated the same procedure for 500, 1000 and 5000 Oe.

In figure 5 we show the results of dc magnetic measurements. Two things are noticeable with respect to the nature of the magnetic transition: (i) irreversible points between the ZFC and FC data and (ii) the temperature of the maximum of the ZFC curve. This temperature of the maximum defined as a spin glass transition decreases with increasing field. The maximum in the data taken with 100 Oe is located at 5.6 K, lower than the maximum in the real part of the ac susceptibility taken with the lowest measured frequency [16]. Again this difference is consistent with our previous conclusion that the transition we have been discussing all along is indeed of a spin glass nature.

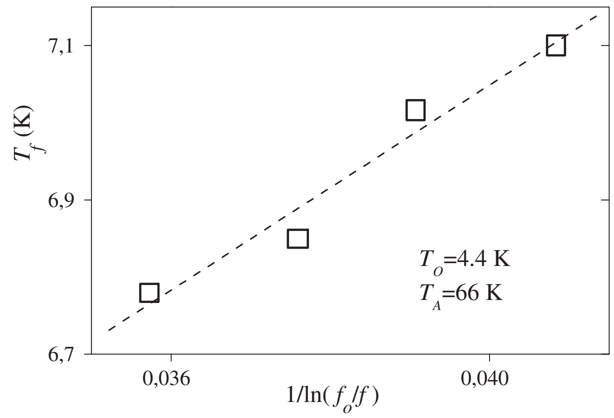

Figure 4. The analysis of the experimental data using the Vogel–Fulcher law. T_f is the position of the maximum of χ' for a given frequency f with $f_0 = 10^{13}$ Hz (see the text).

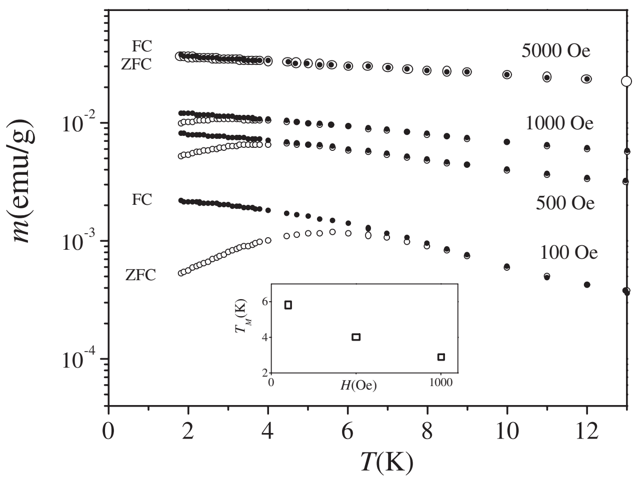

Figure 5. The temperature dependence of the magnetic moment was measured for both ZFC and FC. Inset: the temperature of the maxima in the ZFC curves versus applied field.

5. Relaxation processes and analysis of results

As pointed out in [16], similar ZFC and FC effects can also be found in superparamagnets. Therefore, we have performed some further experiments, which show that the relaxation processes we observed in our sample are genuinely inherent for spin glass behaviour. The sample was initially cooled down to 2 K in zero field. Then the field of 100 Oe was turned on and the magnetization was measured immediately as a function of time. After each measurement, the sample was warmed up to 35 K and then cooled to a target temperature for another relaxation measurement. It took about 1 min to stabilize the temperature and switch the field on after reaching the target temperature. One such $m(t)$ curve is displayed in figure 6. We tried to fit the data using a stretched exponential function [16] as well as an exponential function of the form $\exp(-\Delta/kT)$ originally proposed by Néel for ideal superparamagnets with a single energy barrier [17]. Neither of these functions can fit our experimental data well. We have

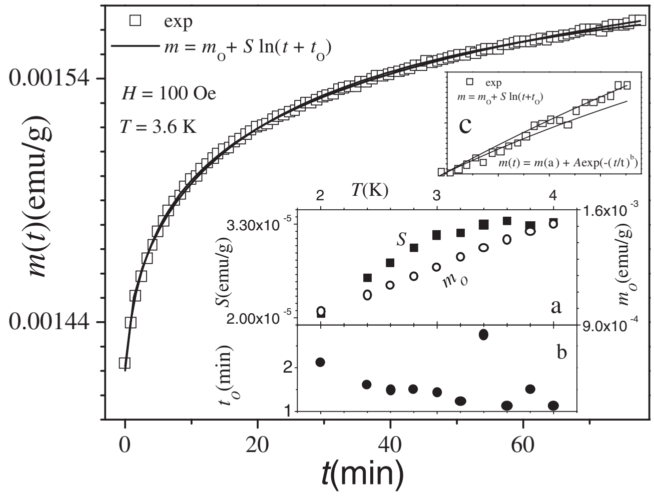

Figure 6. The time dependence of the magnetization under a field of 100 Oe at $T = 3.6$ K. The experimental curve (open squares) is fitted with a function $m(t) = m_O + S\ln(t + t_O)$ (full curve). The temperature dependence of the fitting parameters is given in insets (a), S (full squares) and m_O (open circles), and (b), t_O (full circles). In inset (c) we expand the part of $m(t)$ experimental points where we can see that the stretched exponential function is not able to fit the experimental data (lower curve). For clarity we omit the pure exponential curve, which fits the data much worse.

instead found that the following logarithmic function gives better agreement:

$$m(t) = m_O + S\ln(t + t_O). \quad (5)$$

We note that a similar temperature dependence of $m(t)$ was seen previously in thermoremanent as well as isothermal remanent magnetization processes of $Au_{1-x}Fe_x$ ($x = 0.02$ and 0.07) [18]. In order to fit all our data, we need to introduce t_O. The average value of t_O is 1.6 min, about the same size as the time delay in our measurement after reaching the target temperature. m_O is a linear function of temperature. In equation (5) the constant S, the so-called magnetic viscosity, increases up to 3.6 K (the upper inset of figure 6), i.e. $0.6T_M$, where $T_M = 5.8$ K is the freezing temperature (T_f) for 100 Oe (figure 5). It is to be noted that in $Au_{1-x}Fe_x$ the maximum of S is also found to be located at $0.6T_f$, in good agreement with our data.

6. Discussion and conclusions

We have shown that very dilute U in YRu_2Si_2 has a clear magnetic transition of a spin glass nature. Thus our investigations are in agreement with [6, 7], where the spin glass behaviour was indicated but not confirmed in the U doped $ThRu_2Si_2$ experiments. The effects due to the transition are small, especially the magnetic contributions to the imaginary part of the susceptibility ($\Delta\chi''$) and the corresponding contribution to the resistivity ($\Delta\rho$). Regarding the origin of the spin glass transition, we acknowledge that a possible effect due to the secondary phase is certainly not easy to rule out. However, we believe that, because of the following reasons, the spin glass transition is genuinely due to dilute U moments coupled through the RKKY interactions. First, the fact that we observed a similar feature at low temperatures in all our measurements makes this secondary phase scenario unlikely. In particular, the resistivity would not be very sensitive to such a small impurity phase. Second, the frequency dependence of the transition temperature is consistent with the conventional wisdom of the spin glass transition. However, that the peak in the ac susceptibility is rather broad compared with those seen in other conventional spin glass systems may be due to inhomogeneity of the sample composition. It may well be possible that, after long annealing, it becomes sharper.

In summary, we have performed an extensive study of a low U doped YRu_2Si_2 alloy using several experimental techniques such as ac and dc magnetic susceptibility, dc resistivity and microstructural investigations and we have found a spin glass transition in very dilute U doped YRu_2Si_2. Our conclusion is based on the present knowledge and understanding of this phenomenon and is supported by similar investigations on other systems. This transition is mediated through the RKKY interaction between U moments, which is helped by the very extended nature of the U 5f wavefunction.

Acknowledgments

Work at SungKyunKwan University was supported by Nuclear R & D programs of the Ministry of Science and Technology and the Centre for Strongly Correlated Materials Research.

References

[1] Palstra T T M, Menovsky A A, van der Berg J, Dirkmaat A J, Kes P H, Nieuwenhuys G J and Mydosh J A 1985 *Phys. Rev. Lett.* **55** 2727
[2] Santini P and Amoretti G 1994 *Phys. Rev. Lett.* **73** 1027
Santini P 1998 *Phys. Rev.* B **57** 5191
[3] Amitsuka H and Sakakibara T 1994 *J. Phys. Soc. Japan* **63** 736
[4] Park J-G, McEwen K A and Bull M 2002 *Phys. Rev.* B **65** 094500
[5] Amitsuka H, Kuwahara K, Yoshida T, Tenya K, Sakakibara T, Mihalik M and Menovsky A A 1999 *Physica* B **259–261** 412
[6] Andraka B and Tsvelik A 1991 *Phys. Rev. Lett.* **67** 2886
[7] Kim W W, Steward G R and Andraka B 1994 *Physica* B **199/200** 384
[8] Liu L Z, Allen J W, Seaman C L, Maple M B, Dalichaouch Y, Kang J-S, Torikachvili M S and Lopez de la Tore M A 1992 *Phys. Rev. Lett.* **68** 1034
[9] Park J-G and Coles B R 1994 *J. Phys.: Condens. Matter* **6** 1425
Park J-G 1994 *J. Phys.: Condens. Matter* **6** 3403
Park J-G, Roy S B and Coles B R 1994 *J. Phys.: Condens. Matter* **6** 5937
[10] Očko M, Park J-G and Aviani I 1999 *Physica* B **259–261** 260
[11] Süllow S, Mattheaus C C, Becker B, Snel C E, Schenk A, Nieuwenhuys G J and Maydosh J A 1997 *Physica* B **230–232** 43
[12] Süllow S, Mentink S A M, Mason T E, Buyers W J L, Nieuwenhuys G J, Menkovsky A A and Maydosh J A 1997 *Physica* B **230–232** 105
[13] Landau L and Lifshitz E 1969 *Electrodynamique des Milieux Continus* (Moscow: Mir)
et al Khoder A F and Couach M 1991 *Magnetic Susceptibility of Superconductors and Other Spin Systems* ed R A Hein (New York: Plenum) p 213
[14] Reger J D and Binder K Z 1985 *Physica* B **60** 137
[15] Tholence J L 1980 *Solid State Commun.* **35** 113
[16] Fisher I R, Cheon K O, Panchula A F, Canfield P C, Cherinkov M, Ott H R and Dennis K 1999 *Phys. Rev.* B **59** 308
[17] Néel L 1949 *Ann. Géophys.* **5** 99
[18] Guy C N 1978 *J. Phys. F: Met. Phys.* **8** 1505