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Non-linear susceptibility in heavy fermion compounds $CeRu_2Si_2$ and $Ce_{1-x}Y_xRu_2Si_2$ ($x \leqslant 0.1$)

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Abstract. We report linear and non-linear susceptibility measurements performed on three single crystals of the heavy fermion compound $CeRu_2Si_2$. For the easy magnetization axis (H parallel (||) to the c-axis of tetragonal structure), the linear susceptibility, $\chi_{1\|}$, of $CeRu_2Si_2$ shows the previously reported rounded maximum near 11 K. New results are the existence of a non-linear susceptibility, $\chi_{3\|}$, which shows a shallow dip near 25 K and a peak near 6 K. These two temperatures can be related to T_K and to the saturation of antiferromagnetic correlation lengths, respectively. Similarly $\chi_{3\|}$ is observed in $Ce_{1-x}Y_xRu_2Si_2$ ($x = 0.015$, 0.05 and 0.1). This observation of non-linear susceptibility raises the question of the possible existence of quadrupolar interactions in $CeRu_2Si_2$, like in other classical rare-earth compound.

1. Introduction

The non-linear susceptibility in rare earth intermetallic compounds has been rather well interpreted in terms of quadrupolar interactions present in the systems [1]. Up to now, however, such studies were mostly limited to conventional rare earth systems. Only a few studies of heavy fermion compounds have been reported so far. Among them are reports by two independent groups [2, 3] of somewhat anomalous behaviour of non-linear susceptibility in URu_2Si_2. Both groups claim that their observations suggest that 'itinerant' quadrupolar interactions contribute to the antiferromagnetic (AF) ordering at 17.5 K in this compound. However, the quantitative interpretation of the role of quadrupolar interactions in heavy fermion compounds appear not to be as easy as in more classical rare earth systems. Since, as shown in [1], the proper interpretation of non-linear susceptibility requires a good knowledge of crystal field effects in the system considered, this might be one of problems with heavy fermion compounds. Nevertheless, we believe, it is worth studying non-linear susceptibility in such compounds, especially because either they are close to a magnetic instability or they show anomalous magnetic ordering with tiny ordered magnetic moments. From this point of view, URu_2Si_2 could be a good candidate for such studies since its ordered moment is merely 0.04 μ_B and the nature of its magnetic transition is yet to be understood.

Its homologous compound, $CeRu_2Si_2$ appears to be another good candidate for non-linear susceptibility study, because it shows quite non-trivial magnetic properties at low

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temperatures. Neutron studies [4] have shown the occurrence of short range AF correlations below ~70 K, increasing rapidly below 15 K and saturating below 6 K (still far from any long-range ordering). At low temperatures, these correlations disappear smoothly above the field ($B^* \sim 8$ T) where a pseudometamagnetic transition occurs in the magnetization for H parallel to the c-axis [5, 6]. Recent muon-spin experiments [7] suggest the occurrence of static moments of the order of 10^{-3} μ_B below 1.5–2 K, coexisting with the AF correlations. As far as other properties are concerned, $CeRu_2Si_2$ shows typical non-magnetic heavy fermion behaviours: AT^2 term in the resistivity below about 1 K [8] with $A \sim 0.4$–0.6 $\mu\Omega$cm K^{-2}, rounded maximum near 11 K in the linear susceptibility, which decreases as T^2 below ~4 K to a residual value [5, 6], smooth increase of C/T at low temperature, leading to a saturation value of 350 mJ K^{-2} mol^{-1} at 0.1 K [6]. Only a small anomaly in thermopower below ~2 K [9] and a smooth change of slope in the Hall coefficient R_H versus T^2 around 2 K [10] might be regarded as signatures of the occurrence of the above static moments.

2. Experiments

For these studies, we used three different $CeRu_2Si_2$ single crystals (which we will further label $CeRu_2Si_2$ M1, M2 and M3), and three single crystals of $Ce_{1-x}Y_xRu_2Si_2$ with $x =$ 0.015, 0.05 and 0.1.

All these crystals were cut from rods grown by the Czochralski pulling technique in a tri-arc furnace under purified argon atmosphere; starting materials were of high purity, as described elsewhere [11]. $CeRu_2Si_2$ M1 and the $Ce_{1-x}Y_xRu_2Si_2$ crystals were measured 'as cast', i.e. without further heat treatment after pulling. $CeRu_2Si_2$ M1 is the same crystal as one used in [6] while $CeRu_2Si_2$ M2 and $Ce_{0.985}Y_{0.015}Ru_2Si_2$ are those used for thermal expansion and magnetostriction measurements ([12] and [13], respectively). However, $CeRu_2Si_2$ M2 has been annealed for eight days at 850 °C in 10^{-6} torr before we performed our measurements. Finally, $CeRu_2Si_2$ M3 is a new crystal grown in a new furnace outgased by an ultrahigh vacuum (UHV) system and annealed for eight days at 1100 °C in UHV (10^{-10} torr). According to resistivity measurements performed on small adjacent pieces, the three $CeRu_2Si_2$ crystals differ by their resistivity ratios and residual resistivity values: for the current along the basal plane $\rho_{1.2\ K} = 2.5$, 0.9 and 0.7 $\mu\Omega$cm and $\rho_{290}/\rho_{1.2} = 45$, 92 and 140, for M1, M2 and M3, respectively. Taking into account the AT^2 decrease of ρ below ~1 K, one can easily expect residual resistivities ρ_0 ($\equiv \rho_{15\ mK}$) lower than $\rho_{1.2}$ and ρ_{290}/ρ_0 higher than $\rho_{290}/\rho_{1.2}$ by about 10–20%. For the $Ce_{1-x}Y_xRu_2Si_2$ system, resistivity measurements show that the residual resistivity value increases by 3 $\mu\Omega$cm per percent of Y.

Both linear and non-linear susceptibilities were derived from magnetization, $M(H)$ measured in home-made extraction magnetometers. We used an apparatus of sensitivity 10^{-3} emu with a 7.5 tesla coil, except for $CeRu_2Si_2$ M2 which has been measured using an apparatus of slightly lower sensitivity.

3. Results

According to the field expansion of M up to third order $M = \chi_1 H + \chi_3 H^3/3!$, χ_1 and χ_3 can be derived from plots of M/H against H^2. For most of our samples, these plots show a small upturn of M/H at low field ($H < 2$ T). Some examples are given in figure 1

for $CeRu_2Si_2$ M3. As illustrated in figure 2 by plots of $\Delta M/H$ versus $1/H$, such upturns are well accounted for by the presence of a tiny ferromagnetic impurity of the order of 10^{-3} μ_B. Apart from that, it was always possible to get a linear behaviour in M/H against H^2 plots for H higher than 2 tesla and to define the intercept and the slope of a straight line at all temperatures. However, for $H \parallel c$, the field range for our analysis becomes narrow at low temperatures because M/H starts to exhibit an upturn above 4 T or less due to the pseudometamagnetic transition. For $CeRu_2Si_2$, it is possible that a small part of this pseudometamagnetic upturn is included in the value of χ_3, thus our χ_3 being a little overestimated at the lower temperatures. This difficulty disappears with Y-doping, since the value of B^* increases almost exponentially with x ($B^* \sim 9.4$, 13 and 20 T, for $x = 0.015$ [13], 0.05 [14] and 0.1 [15], respectively. For La-doped systems, however, it becomes impossible to define a value of χ_3 at lower temperatures because M/H shows an increase in even low field clearly due to the pseudomagnetic transition ($B^* \sim 5.3$ T for 5% La) [6, 13, 14]. The transverse susceptibility was measured only in a few cases when the mass of the crystals were large enough to lead to a good precision with our extraction magnetometers. For $H \perp c$-axis, no metamagnetic transition occurs [5] and, except for the low field upturn, M/H against H^2 plots are straight lines in all the field range studied.

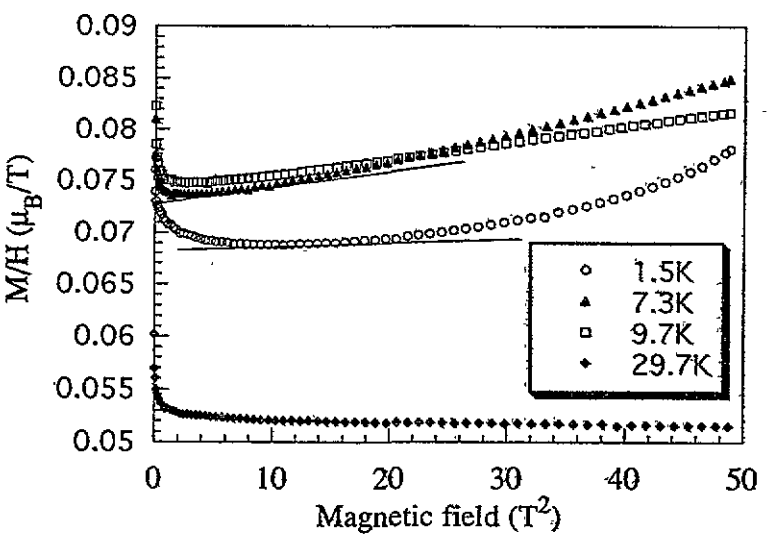

Figure 1. M/H versus H^2 for $CeRu_2Si_2$ M3. Lines under 1.5 and 7.3 K data are our estimation of $M = \chi_1 H + \chi_3 H^3/3!$ Curvatures seen at higher field are due to the pseudometamagnetic transition of $CeRu_2Si_2$. For 9.7 and 29.7 K data where the metamagnetic transition disappears, M/H is linear to H^2 up to 7 teslas apart from a low-field region (see the text).

The temperature dependence of the linear and non-linear susceptibilities are plotted in figures 3 and 4. When measured with $H \parallel c$, they are labelled as $\chi_{1\parallel}$ and $\chi_{3\parallel}$. With H applied along the basal plane, corresponding susceptibilities are noted as $\chi_{1\perp}$ and $\chi_{3\perp}$. Note that the variations of $\chi_{1\parallel}$ reported here for $CeRu_2Si_2$ M1 are almost identical to those in [6], because we used the same magnetization data. Instead of taking the intercept M/H against H^2 plots as described above, the values of $\chi_{1\parallel}$ reported in [6] were deduced from $\partial M/\partial H$ against H plots, neglecting the $\chi_{3\parallel}$ term. As we saw the low-field upturn in M/H against H^2 plots, $\partial M/\partial H$ against H plots also showed that the low-field data points cannot

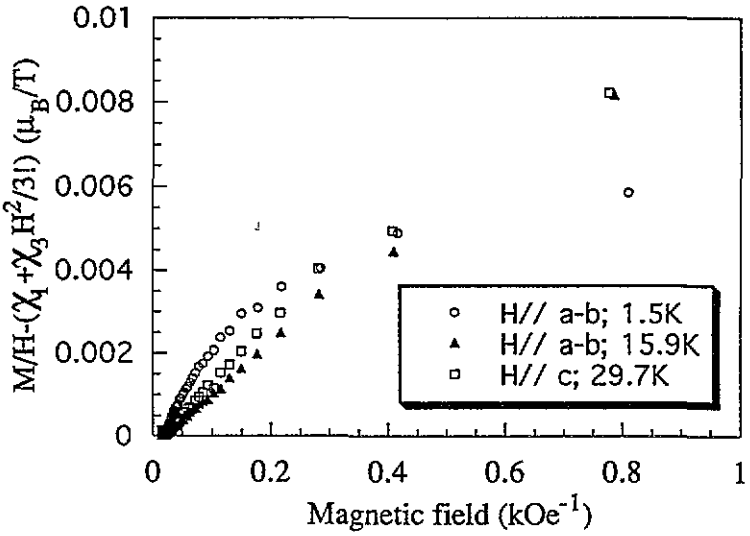

Figure 2. $\Delta M/H$ $(= M/H - (\chi_1 + \chi_3 H^2/3!))$ against $1/H$ for $CeRu_2Si_2$ M3. From the graph, we can roughly estimate that the upturn at low field in M/H against H^2 plots is due to 10^{-3} μ_B/f.u. of ferromagnetic impurities in it.

be taken into account to define the susceptibility. We will now describe the main features observed in our data.

3.1. Variations of $\chi_{1\parallel}$ and $\chi_{1\perp}$

In figure 3, we show $\chi_{1\parallel}$ for $CeRu_2Si_2$ M1 and three Y-doped alloys. As reported previously, it is noteworthy that there appears a rounded maximum, followed at lower temperatures by a decrease of less than 10% to a residual value. For $CeRu_2Si_2$, this maximum occurs near 11 K. It is interesting to note that the crystals of better quality, $CeRu_2Si_2$ M2 and M3 show a somewhat higher value of $\chi_{1\parallel}$ than $CeRu_2Si_2$ M1 ($\chi_{1\parallel} \sim 7.45 \times 10^{-2}$ μ_B T^{-1} instead of 7.07 at the maximum). However, the temperature of this maximum, $T_{max\,1}$ is the same for the three crystals, thus we choose to plot only one curve, for clarity. In $Ce_{1-x}Y_xRu_2Si_2$, one can see a decrease in the maximum value of $\chi_{1\parallel}$ and an increase in the value of $T_{max\,1}$ on increasing x. As far as these variations of the maximum are concerned, the effects of Y-doping are more or less equivalent to the application of an external pressure on $CeRu_2Si_2$ [16] where the increase of $T_{max\,1}$ has been related to the increase of the characteristic temperature, or Kondo temperature T_K of the system [16, 17], following a scaling behaviour and leading to a Grüneisen parameter of 180.

To show the anisotropic nature of magnetic susceptibility, we also present $\chi_{1\perp}$ for a 10% Y-doped sample in figure 3. For comparison, we had found $\chi_{1\parallel}/\chi_{1\perp}$ close to 20 for $CeRu_2Si_2$ M1 previously [18]. Thus, the anisotropy ratio, $\chi_{1\parallel}/\chi_{1\perp}$ decreases with replacing Ce by Y (while it increases with doping La).

As previously seen, $\chi_{1\perp}$ shows a rather broad maximum where $\chi_{1\parallel}$ has a relatively clear feature. This maximum in $\chi_{1\perp}$ is found to be present in all samples we studied.

3.2. Non-linear susceptibilities ($\chi_{3\parallel}$ and $\chi_{3\perp}$)

To show more clearly interesting aspects of χ_3, in figure 4 we present $\chi_{3\parallel}$ along with $\chi_{1\parallel}$ for $CeRu_2Si_2$ M1. While $\chi_{1\parallel}$ has a modest maximum at 11 K and a Curie–Weiss behaviour

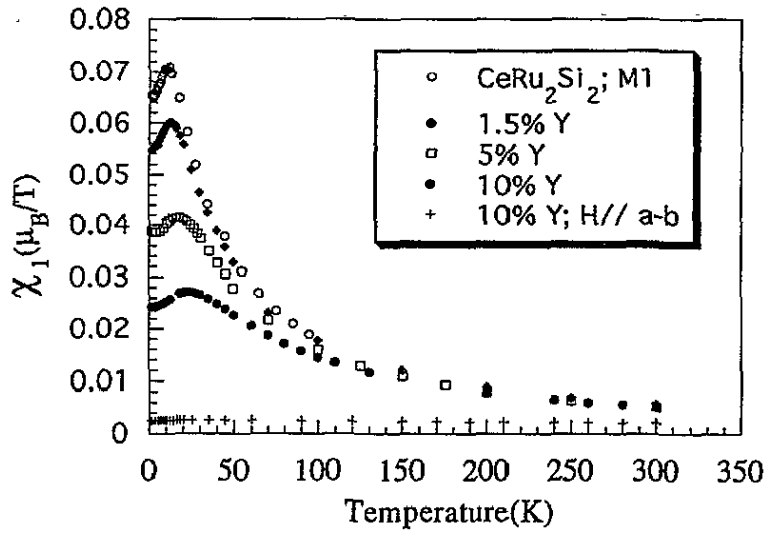

Figure 3. Linear susceptibilities, $\chi_{1\|}$, for the samples. $\chi_{1\perp}$ is also included in the figure for 10% Y-doped sample.

above 30 K (see [5] and [6] for details), $\chi_{3\|}$ has a peak centred at 6 K, becomes negative above 20 K with a shallow dip and reaches zero around 60 K (see figure 4). Above 60 K, $\chi_{3\|}$ is almost temperature independent. Apart from the fact that $\chi_{3\|}$ shows rich features compared with $\chi_{1\|}$, it is interesting to note that $\chi_{3\|}$ becomes very much temperature dependent below 60 K where inelastic neutron experiments [4] saw the development of antiferromagnetic correlations. This point will be further addressed later.

In figure 5(*a*), we show $\chi_{3\|}$ for all three $CeRu_2Si_2$ single crystals and $\chi_{3\perp}$ for $CeRu_2Si_2$ M3 sample; it is to be stressed that $\chi_{3\|}$ is not sample dependent. Apart from small differences from one sample to another, they all agree that $\chi_{3\|}$ has a peak around 6 K and becomes

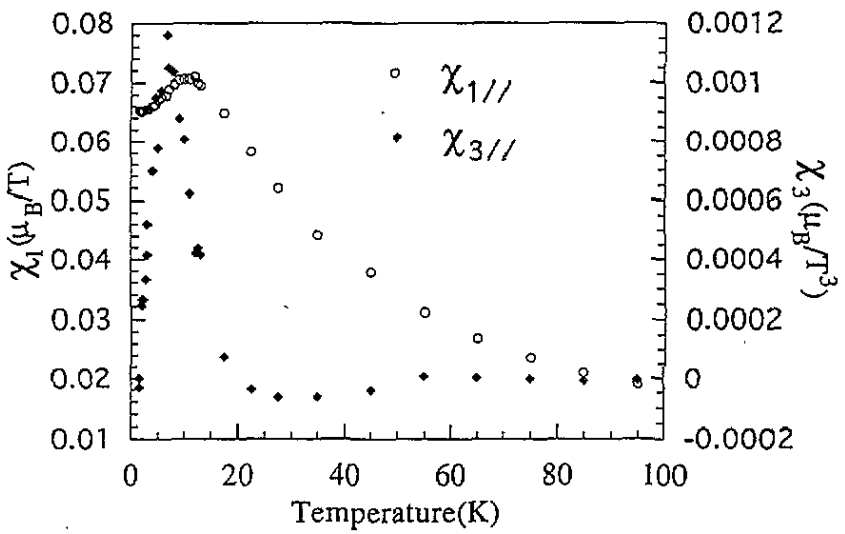

Figure 4. χ_1 and χ_3 are presented for $CeRu_2Si_2$ M1.

negative above 20 K; the small differences, we believe, are due to the quality of samples. It is also noteworthy that like χ_1, χ_3 has also shown a strong anisotropic nature; $\chi_{3\perp}$ is almost zero and tends to be negative below around 6 K.

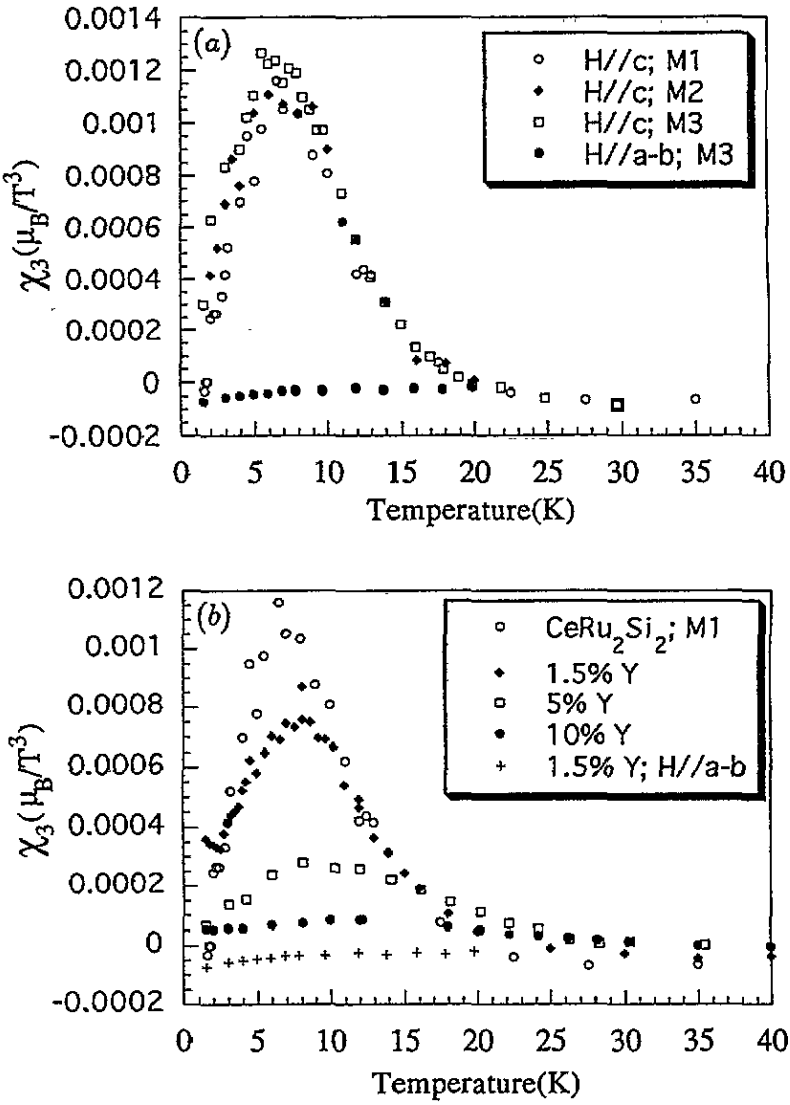

Figure 5. (*a*) $\chi_{3\parallel}$ data are presented for three $CeRu_2Si_2$ single crystals along with $\chi_{3\perp}$ for $CeRu_2Si_2$ M3. (*b*) $\chi_{3\parallel}$ and $\chi_{3\perp}$ data for Y-doped alloys. Data for $CeRu_2Si_2$ M1 are included for comparison.

With doping of Y, the non-linear susceptibility becomes attenuated while the peak temperature shifts towards higher temperature (see figure 5(*b*)). Roughly speaking, there seems to be a relation between the peak in $\chi_{3\parallel}$ and the maximum in $\chi_{1\parallel}$ (see table 1). Compared with $\chi_{1\parallel}$ in figure 3, we would like to stress that although $\chi_{1\parallel}$ at the maximum temperature is reduced about 1/7 with 10% Y doping, $\chi_{3\parallel}$ for 10% Y-doped alloy at

the peak temperature is almost 5% of that for $CeRu_2Si_2$ which is an indication of how sensitive the non-linear susceptibility is to alloying. About the extremely attenuated non-linear susceptibility under Y doping, we may recall that the Kondo temperature is known to increase from 24 K for $CeRu_2Si_2$ to 50 K for 10% Y doped case [19]. Therefore, the antiferromagnetic correlations seen in $CeRu_2Si_2$ are expected to be extremely small in the Y-doped samples. All non-linear susceptibilities, $\chi_{3\|}$, seem to become negative above almost the same temperature (around 20 K), allowing small differences. It is also to be noted that the anisotropy ratio in χ_3 for a 10% Y-doped alloy is about 1 at the lowest temperature while it is near to 10 in χ_1.

Table 1. Characteristic temperatures of linear $\chi_{1\|}$ and non-linear $\chi_{3\|}$ susceptibilities of $CeRu_2Si_2$ and $Ce_{1-x}Y_xRu_2Si_2$ alloys for easy magnetization direction.

x	$T_{max\,1}$ temperature of the maximum of $\chi_{1\|}$ (K)	$T_{max\,3}$ temperature of the maximum of $\chi_{3\|}$ (K)	$T_{min\,3}$ temperature of the minimum of $\chi_{3\|}$ (K)
0	11	6	25–30
0.015	12.5	7.5	35
0.05	18	9	45
0.1	25	13	45

4. Discussion

In order to understand the behaviour of χ_3 in $CeRu_2Si_2$ (particularly $\chi_{3\|}$), it is worth comparing them with other reported data [2, 3]. In each case, only χ_3 along the easy magnetic direction show somewhat interesting features. In U_2Zn_{17} and $U_{0.95}Th_{0.05}Pt_3$, non-linear susceptibility changes its sign from small and negative above T_N to positive (and much larger) below T_N. The authors of [2] conclude that these compounds follow the expected response for a normal antiferromagnet. Although a similar change in sign occurs in the $\chi_{3\|}$ of $CeRu_2Si_2$ near 20 K, however, it is to be noted that this temperature has nothing to do with magnetic ordering temperature.

Since URu_2Si_2 is another heavy fermion compound as well as an homologous compound, it may be interesting to compare non-linear susceptibility for the two. In URu_2Si_2, the c-axis is the magnetic easy axis. For URu_2Si_2, $\chi_{3\|}$ exhibits a peak in the vicinity of T_N (=17.5 K) while $\chi_{3\perp}$ is small, negative and almost temperature independent over the temperature range [2, 3]. As to the peak, it should be realized that a reconstruction of the Fermi surface occurs at the same temperature as the peak, resulting from the opening of a gap in the excitation spectrum and also giving rise to anomalies in other properties (see references given in [2] and [3]). From their studies, both groups conclude that this behaviour is related to 'itinerant' quadrupolar interactions. However, it is important to mention that recently one group [20] has shown using polarized neutron techniques that the 17.5 K antiferromagnetic transition is unlikely due to quadrupolar interactions.

Concerning $CeRu_2Si_2$, it seems possible to connect its non-linear susceptibilities to other properties. In our data, an interesting point is the maximum of χ_3 occurring near 6 K. Neutron studies [4] have already shown that the antiferromagnetic correlation length becomes temperature independent below about this temperature, after a rapid increase below

10–15 K. A temperature of 5–6 K corresponds also to some anomalous behaviours in transport properties: the value of the pseudometamagnetic field B^*, when defined by the maximum of the magnetoresistance with the current along the c-axis [21], shows a large increase between 20 K ($B^* \sim 1$ T) and 6 K ($B^* \sim 7$ T) and saturates around 7.5 T below about 5 K. Similarly, the latter value of B^* can be detected in the variation of the thermopower (S) versus magnetic field only below about 6 K [9] (minimum of $S(B)$).

An interesting question will be what could happen at the 'ordering' temperature of 1.5–2 K detected by muon-spin experiments [7]. As recalled in the introduction, no anomalous behaviour is seen at this temperature in other physical properties. Related to this, it is interesting to note the rapid decrease of $\chi_{3\|}$ below 6 K (which could be more steep since we are likely to overestimate this quantity at lower temperatures as discussed in the previous section). For $CeRu_2Si_2$, the value of $\chi_{3\|}$ falls almost to zero near 1.5 K (our lowest temperature). Thus it appears that more work is needed at low temperature to check whether ordering reported in [7] has some implications on $\chi_{3\|}$. Since we also saw $\chi_{3\|}$ of $Ce_{1-x}Y_xRu_2Si_2$ alloys decrease rapidly below the maximum, however one would expect some complications in drawing a correlation between χ_3 of $CeRu_2Si_2$ below 2 K and the ordering temperature observed by μSR.

The high temperature behaviours of $\chi_{3\|}$ are found to be difficult to explain. Turning back to URu_2Si_2 for comparison, the data in [3] show that in this compound $\chi_{3\|}$ is almost constant above the peak at T_N up to 50 K and then decreases upon heating, reaching zero near 200 K. Myako *et al* [3] related this behaviour to a crystal field effect, in good agreement with thermal expansion studies. In $CeRu_2Si_2$, however the crystal field level scheme is 0–220–1000 K as inferred from specific heat data [19], while the variation of the volume thermal expansion coefficient above 100 K is better fitted by a splitting between the ground state and the first excited level of 280 K instead of 220 K [22]. Therefore it seems hard to find any connection between this level scheme and the minimum of $\chi_{3\|}$ near 25–30 K. Perhaps this temperature can be better compared with the Kondo temperature T_K of $CeRu_2Si_2$, which has been estimated to 24 K from specific heat measurements [19], confirmed by the neutron experiments [4]. The minimum in $\chi_{3\|}$ is shifted to higher values with Y doping (up to ~45 K for $x = 0.1$); accordingly substitutions of Y for Ce is known to increase the value of T_K. However, this remains only qualitative. Moreover, the small but sharp increase in $\chi_{3\|}$, which is not shown in the paper, above this 45 K minimum for $x = 0.1$, is yet to be explained.

Finally, we would like to address the question of whether quadrupolar interactions play a role in $CeRu_2Si_2$, since the existence of non-linear susceptibility in some rare-earth intermetallic compounds has been often interpreted to be due to quadrupolar interactions [1]. As mentioned briefly in the introduction, one problem is that for heavy fermion compounds the knowledge of crystal field effect is lacking because of the strong hybridization of the f-electrons. Although the problem is slightly relieved in $CeRu_2Si_2$; crystal field splitting can be decided using specific heat data [19], a coherent picture like those in [1] for classical systems is unlikely to emerge.

In considering whether quadrupolar interactions are present in a certain system, elastic constants and magnetostriction have also been used along with non-linear susceptibility. For example, $C^{\gamma}(=C_{11}-C_{12})$ mode becomes extremely softened in the paramagnetic state of magnetic intermetallic compounds where quadrupolar interactions are known to exist. Magnetostriction in quadrupolar systems is relatively large, although it is not straightforward to regard large magnetostriction as an indication of quadrupolar interactions. For $CeRu_2Si_2$ both elastic constants [23, 24] and magnostrictions [25] have been measured, but mainly in order to study the pseudometamagnetic transition occurring at B^* (=7.6 T

at low temperature). At this pseudometamagnetic transition, isothermal longitudinal elastic constants get softened up to 50% at 60 mK, while isothermal magnetostriction shows a peak which reaches a value of 10^{-3} near 0.1 K. Despite that few theoretical models have been proposed of the pseudometamagnetic transition (see references given in [24] and [25]), however they seem to us not to be satisfactory in explaining both elastic constants and magnetostrictions consistently. We think it is important that antiferromagnetic correlations disappear above B^*.

In conclusion, we would like to note that in most temperature ranges studied $CeRu_2Si_2$ does not have long-range magnetic order but is expected to be magnetic with a very tiny ordered moment only below 1.5 to 2 K [7]. It is thus worth emphasizing that in cases like heavy fermion compounds where dipole magnetic moments are screened rather well by conduction electrons through Kondo screening, it may be important to consider next order interactions, however small they are.

5. Conclusion

We have observed several interesting points in the non-linear susceptibility of $CeRu_2Si_2$ and $Ce_{1-x}Y_xRu_2Si_2$ dilute alloys for easy magnetic direction. For the pure compound, the temperature dependence of $\chi_{3\|}$ can be related to the Kondo temperature and another temperature below which the antiferromagnetic correlation length saturates. Although quadrupolar interactions might be responsible for the behaviour of $\chi_{3\|}$, the situation is expected to be somewhat complicated in such heavy fermion systems and the origin of the non-linear susceptibility we have observed in $CeRu_2Si_2$ remains an open question.

Acknowledgments

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