^{29}Si NMR study of the heavy fermion system CeRuSi

Journal of Physics: Conference Series **200** (2010) 012173 doi:10.1088/1742-6596/200/1/012173

^{29}Si NMR study of the heavy fermion system CeRuSi

Rajib Sarkar‡, Michael Baenitz, † Julian Sereni, and Cristoph Geibel

Max-Planck Institute for Chemical Physics of Solids, 01187 Dresden, Germany
†Divisin Bajas Temperaturas, Centro Atmico Bariloche, Comisin Nacional de Energia Atmica, cc 439, 8400 San Carlos de Bariloche, Argentina

E-mail: rajibsarkarsinp@gmail.com

Abstract. A recent investigation of the low temperature properties of the paramagnetic heavy fermion system CeRuSi revealed an anomalous decrease of the coefficient $\frac{C}{T}$ below 6 K, indicating a second low energy scale well below the Kondo temperature of about 50 K. We started an investigation of this unusual behavior using the local probe ^{29}Si-NMR. Here a preliminary ^{29}Si field sweep NMR study in the temperature 4.2-200 K is reported. ^{29}Si-NMR powder spectra obtained at 72.9 MHz could be simulated and shift components $^{29}K_{ab}(T)$ and $^{29}K_c(T)$ were determined. The shift along the basal plane $^{29}K_{ab}(T)$ shows the expected behavior with a Curie-Weiss dependence at high T merging in a constant value below 15 K. In contrast the shift along the tetragonal axis reveals an anomalous T dependence below 15 K where $^{29}K_c(T)$ drops by a factor of 2, confirming the presence of a further low T energy scale. We suspect this drop to be related to the anomalous decrease of $\frac{C}{T}$ below 6 K and discuss possible mechanism.

1. Introduction

In contrast to the thorough investigation of $CeRu_2Si_2$ in the past 30 years, only very few results have been published on the related compound CeRuSi [3][4] , although it presents some interesting features. It crystallizes in the CeFeSi [2] structure type, which differs from the structure of the new RFeAsO (R: rare earth) high T_c superconductors only by removing the O atoms. This quasi-2D structure should enhance quantum fluctuations conductive to the observation of unusual properties. A detailed investigation confirmed a paramagnetic heavy fermion state with a Sommerfeld coefficient $\gamma = 175\ \frac{\mathrm{mJ}}{\mathrm{molK}^2}$ and a Kondo temperature $T_K \approx 50K$, but evidenced an unusual decrease in $\frac{C}{T}$ below 6 K [1]. To investigate the origin of this decrease we started a microscopic study using ^{29}Si (I=1/2) NMR as a local probe. Being a I=1/2 nuclei, no quadrupolar splitting has to be taken into account which makes the ^{29}Si-NMR spectra easier to analyse. The advantage of NMR is that it senses the local susceptibility in a lattice and the results are generally

‡ Author to whom all correspondence should be made.

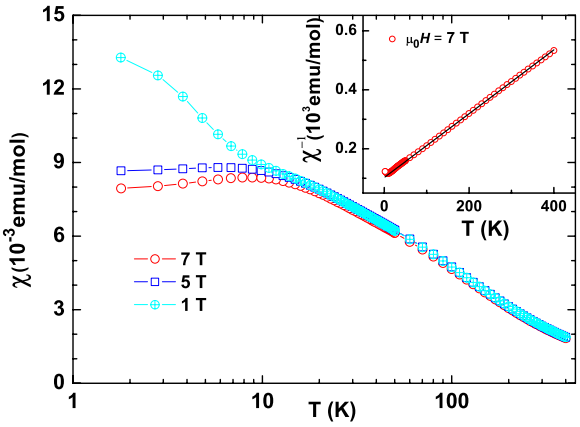

Figure 1. T dependence of the bulk susceptibilty in various fields. The inset shows the CW plot for $\mu_0 H$ =7 T.

not effected by magnetic impurities. Moreover even for a polycrystalline samples with random grain orientation NMR is able to probe the anisotropy of the magnetism. In this paper we report the ^{29}K shift in the temperature range 4.2-200K.

2. Experimental

The single-phase polycrystalline powder sample of CeRuSi was prepared as mentioned in [1]. The powder was fixed in the paraffin to ensure a random orientation. ^{29}Si NMR measurements were performed with a standard pulsed NMR spectrometer (Tecmag). Above 200 K the signal could not be resolved. The field sweep NMR spectra were obtained by integrating the echo in the time domain and plotting the resulting intensity as a function of the field. $^{29}K(T)$ was determined respective to the ^{29}Si NMR line obtained from the duran quartz tube.

3. Results

Fig. 1 shows the magnetic susceptibility $\chi(T)$ of a bulk sample at the field of 7 T, 5 T and 1 T in the temperature range 400-1.8 K. At 1 T there is a upturn below 10 K, whereas at higher fields $\chi(T)$ exhibits a broad maximum followed by a saturation. The Curie Weiss-plot is shown as an inset. The estimated Weiss temperature θ and the effective magnetic moment μ_{eff} from the CW fit, are -90 K and 2.69 μ_B respectively. The Weiss temperature indicates a Kondo temperature T_K ≈45 K, estimated as $T_K = \frac{\theta}{2}$ [6]. The determined moment of 2.69 μ_B is close to the teoretical value for $Ce^{3+}(\mu_{\text{eff}} = 2.54\mu_B)$. The observed field dependence in the $\chi(T)$ is likely due to paramagnetic impurities or foreign phases, since it is sample dependent.

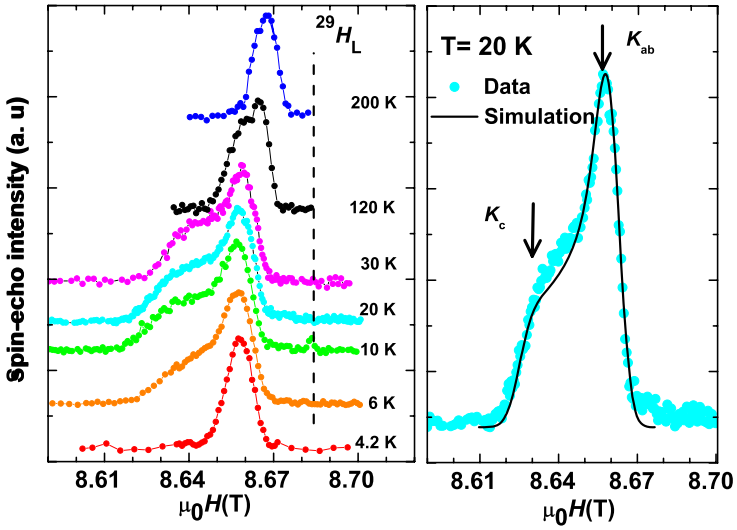

Figure 2. ^{29}Si field-sweep NMR spectra at ν=72.9 MHz and at different temperatures. Vertical dashed line indicates the Larmor field obtained from a reference compound (left). Typical powder spectrum at T=20 K together with the simulation (Right).

^{29}Si field sweep NMR spectra as a function of temperature are shown in Fig. 2. At high temperature one single ^{29}Si NMR line was found as expected from the crystal structure. Upon lowering the temperature it develops a large anisotropy at the low field side as commonly observed in anisotropic systems. Surprisingly, below 10 K the anisotropy reduces again and at 4.2 K the broad spectrum has merged to a single line. The Fig. 2 (right) shows the theoretical simulation at 20 K together with the experimental data. The line shape is typical for a powder pattern from a spin I=1/2 nucleus in a tetragonal structure. The spectra were fitted consistently in the whole temperature range and from there we determined the Knight shift components $K_{\rm ab}(T)$ and $K_{\rm c}(T)$ corresponding to the $H\perp c$ and $H\|c$ directions. The arrows in the right part of Fig.2 indicate $K_{\rm ab}(T)$ and $K_{\rm c}(T)$ contribution of the Knight shift. The vertical dashed line in the left part of Fig.2 indicates the Larmor field $^{29}H_L$. The small additional line showing up in the 10 K spectra at the field 8.682 T originates from the silicon of the sample container.

Fig. 3 shows the temperature dependence of the different shift component $^{29}K_{\rm ab}$, $^{29}K_{\rm c}$ and $^{29}K_{\rm iso}$. It is seen that $K_{\rm ab}(T)$ slowly increases with lowering the temperature and show a shallow maximum at around 12 K as expected from $\chi_{\rm iso}$. In contrast $K_{\rm c}(T)$ increases much more with decreasing T, but below $T^* = 15$ K, it shows an unexpected drop by a factor of 2. Thus the anisotropic shift of this compound is temperature sensitive. If we compare the shift of $^{29}K_{\rm c}(T)$ and $^{29}K_{\rm ab}(T)$ at T^*, it is seen that the anisotropy of the transferred field is really important here and $K_{\rm c}(T)$ is 3 times larger than $K_{\rm ab}(T)$. In the Fig. 3 we have also shown the temperature dependence of the

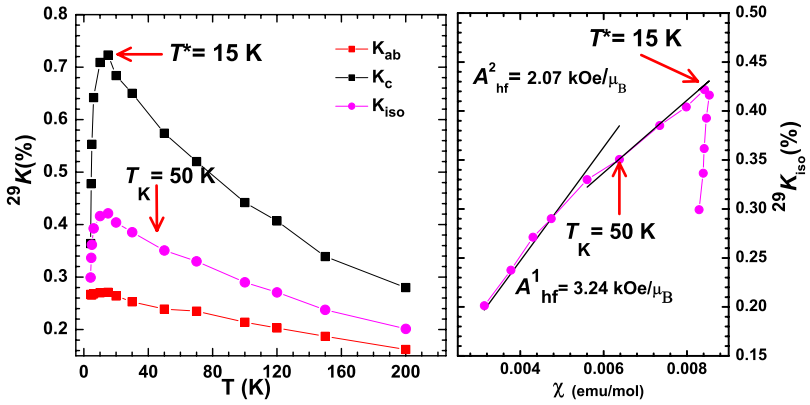

Figure 3. Temperature dependence of $^{29}K_{\rm ab}$, $^{29}K_{\rm iso}$ and $^{29}K_{\rm c}$ (left). $^{29}K_{\rm iso}$ vs. susceptibility plot (right).

$^{29}K_{\rm iso}(T)$, calculated from the relation $^{29}K_{\rm iso} = \frac{1}{3}{}^{29}K_{\rm c} + \frac{2}{3}{}^{29}K_{\rm ab}$. For an estimation of the hyperfine coupling constant $K_{\rm iso}$ is plotted as a function of χ, where χ is the bulk susceptibility. We assume that $\chi = \chi_{\rm iso}$, meaning that there is no alignment or texture in the CeRuSi powder sample. Two changes are visible in the slope of the $\chi_{\rm iso}$ vs. $K_{\rm iso}(T)$ plot. The first weak change occurs at around 60 K, which corresponds roughly to T_K. Such changes of slope near T_K are quite common in Kondo lattice systems. Below $T^* = 15$ K, the slope of $K_{\rm iso}(T)$ vs. $\chi_{\rm iso}$ changes completely, which can be traced back to the drop of $K_{\rm c}(T)$ below T^*. However this is likely an artifact since in such analysis the anisotropy of χ is not taken into account, but this anisotropy becomes large below T^*. The estimated hyperfine coupling constants are $(A^1_{hf}=)$ 3.24 kOe/μ_B above 60 K and $(A^2_{hf}=)$ 2.07 kOe/μ_B for $T^* < T < 60$ K, respectively. The reported A_{hf} for the system $CeRu_2Si_2$ is about 0.91± 0.06 kOe/μ_B, which is less compared to the present system [5].

4. Discussion and summary

The drop in $^{29}K(T)$ is reminiscent of the $\frac{C}{T}$ behaviour. Remarkably, in the concept of heavy quasi particles the relation $K \sim \frac{C}{T} \sim N(E_F)$ is expected well below the Kondo temperature ($T \ll T_K$) where $N(E_F)$ is the density of states at the Fermi level. Therefore the shift should tracks $\frac{C}{T}$, which is seems to be partially the case here. This would suggest the opening of a pseudo gap in $N(E_F)$ at low temperatures. On the other hand the drop in K_c at low T is even larger than that in $\frac{C}{T}$, while the decrease in K_{ab} is only very small. This strong anisotropy cannot be explained by a simple argument based on a reduction in $N(E_F)$. It suggest a strong anisotropy in the T dependence of the susceptibility at low T, i.e. a strong drop in χ_c but almost no decrease in χ_{ab}. In order to be compatible with the bulk susceptibility results, one has then to assume either a strong

texture in the samples used for the susceptibility measurements such as to select the basal plane susceptibility. This is unlikely because of the good reproducibility between different samples. Or one needs $\chi_{\rm ac} < \chi_{\rm ab}$ since then $\chi_{\rm iso}$ is completely dominated by χ_{ab}. Unfortunately, because of the strong peritectic formation of CeRuSi [1], single crystals large enough for the determination of the anisotropy of $\chi(T)$ are yet not available. Therefore this has to be left for future studies. In this context one might argue that a pronounced decrease of χ_c (and thus K_c) below 15 K is due to the crystal field effect resulting in a small c axis moment of a $\Gamma 7$ ground state doublet. However one would then expect the decrease of χ_c to start at a much higher temperature, unless the first excited CEF level is at an energy < 30 K, which is incompatible with present specific heat data [1]. Further on the Kondo interaction with $T_K \approx 50$ K should lead to a mixing of all CEF levels. Thus such a simple mechanism is rather unlikely, and the decrease has to be attributed to some correlation effects. A possible explanation is the onset of two-dimensional antiferromagnetic correlations, since the decrease of K_c occurs in the T -range where AF-ordering was observed in the isostructural compounds CeCoSi and CeCoGe [7]. In a purely localized antiferromagnetic S = 1/2 square lattice systems, one expect a pronounced decrease in $\chi(T)$ below $T = J/k_B$ where J is the exchange parameter between localized spins [8]. Therefore we now started measurements of the relaxation time which could provide information on such correlation effects.

In summary, our investigation of CeRuSi using the local probe ^{29}Si NMR evidenced a strong temperature dependence of the anisotropy of the shift. The shift along the basal plane $^{29}K_{ab}$ shows the expected behavior with a Curie-Weiss dependence at high T merging in a constant value below 15 K. In contrast the shift along the tetragonal axis reveals an anomalous T dependence below 15 K where $^{29}K_c$ drops by a factor of 2, confirming the presence of a further low T energy scale as already inferred from specific heat data [1]. We propose this drop to be related to the anomalous decrease of $\frac{C}{T}$ below 6 K and discuss two dimensional antiferromagnetic correlations as a possible origin. Therefore this feature deserves further investigations, especially with microscopic methods.

References

[1] J. G. Sereni et al. submitted to J. Phys.: Cond. Mat.
[2] R. Welter et al., J. Alloys and Comp., 189 (1992) 49.
[3] B. Chevalier et al., Phys. Rev. B 77 (2008) 014414.
[4] L. Rebelsky et al., J. Appl.Phys. 63 (1988)3405.
[5] Y. Kitaoka et al., J. Phys. Soc. of Jpn. 54 (1985)3236.
[6] H.R. Krishna-murthy et al., Phys. Rev. Lett. 35 (1975) 1101.
[7] B. Chevalier et al., Physica B 378-380 (2006) 795
[8] J.K. Kim, M. Troyer, Phys. Rev. Letter 80 (1998) 2705