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An electron paramagnetic resonance study of the heavy-fermion system URu_2Si_2

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Abstract. Gd EPR measurements on the heavy-fermion system URu_2Si_2 are reported. In agreement with NMR results, we observe a heavy-fermion behaviour for 20 K $< T <$ 60 K and the opening of a spin gap below $T_N = 17.5$ K. Our data reveal an almost constant dynamical susceptibility for $T_N < T <$ 40 K, followed by a Curie–Weiss-like decrease for higher temperatures. The temperature dependence of the magnetic relaxation rate is calculated and compared with that derived from neutron scattering experiments.

1. Introduction

URu_2Si_2 is a well characterized [1, 2, 3] heavy-fermion system (HFS) in which superconductivity coexists with long-range antiferromagnetic (AFM) order [4]. Extrapolation of the Sommerfeld coefficient γ from the paramagnetic regime to $T = 0$ K yields a value of $\gamma = 180$ mJ K^{-2} per formula unit [1]. URu_2Si_2 undergoes an AFM transition at $T_N = 17.5$ K and becomes superconducting below $T_c = 1.2 \pm 0.4$ K. The ordered moment is extremely small and amounts to 0.03 μ_B at lowest temperatures [4, 5]. According to these unusual properties URu_2Si_2 has been characterized as a heavy-fermion band magnet [6].

URu_2Si_2 has been studied using nuclear magnetic resonance (NMR) in full detail [7, 8, 9]. Consistently it has been found that the (nuclear) longitudinal relaxation rate $1/T_1$ is almost constant for $T > 60$ K, decreases linearly from 60 K to 20 K ($(T_1T)^{-1} = 0.28$ s^{-1} K^{-1}), is strongly suppressed below T_N and levels off linearly with $(T_1T)^{-1} = 0.04$ s^{-1} K^{-1}. The strong decrease of T_1^{-1} below T_N provides experimental evidence for the opening of a spin gap in the AFM phase. Also an electron paramagnetic resonance (EPR) study has been performed on URu_2Si_2 using Er^{3+} as an EPR probe [10]. The experiments were performed for $T < 5$ K only and revealed no change of the g-shift at the superconducting phase transition temperature, and only a slight enhancement of the width of the absorption line just below T_c.

A number of neutron scattering experiments have been performed to study magnetic relaxation rates and magnetic excitations in the AFM state [11, 12, 13]. A single-crystal study revealed that a broad and slightly inelastic line (half-width at half-maximum $\Gamma \approx 3$ meV, resonance frequency $\Delta \approx 2.5$ meV for $T > T_N$) changes into a well defined magnetic excitation ($\Gamma \approx 0.5$ meV, $\Delta \approx 4.5$ meV below T_N) [12]. A time-of-flight study of polycrystalline URu_2Si_2 yielded a Korringa-like quasielastic line $\Gamma \approx 3$ meV (30 K) ($T_N < T <$ 50 K) and a dominating inelastic line with $\Delta = 6$ meV and $\Gamma = 1.5$ meV for $T < T_N$ [13].

Here we report on EPR measurements on Gd-doped URu_2Si_2 for temperatures 4.2 K $<$ T $<$ 290 K, focusing especially on the dynamical susceptibility and on the temperature dependence of the magnetic relaxation rate. Our data reveal, in accordance with NMR results [7, 8], heavy-fermion-liquid behaviour for 20 K $<$ T $<$ 60 K and the opening of a spin gap below T_N. Finally we calculate the temperature dependence of the magnetic relaxation rate and compare it with that observed in inelastic neutron scattering experiments.

2. Experimental results and discussion

The Gd-doped $U_{1-x}Gd_xRu_2Si_2$ samples were prepared by arc melting together metals of nominal 4N purity and suction chill casting into a copper mould to produce square-cross-section rods of 0.3 cm $\times$ 0.3 cm $\times$ 4 cm. Samples with Gd concentrations of x = 0.002, 0.005, 0.01, 0.02, and 0.05 were prepared. They were examined metallographically and by x-ray diffraction, and appeared to be single-phase material. URu_2Si_2 crystallizes in the $ThCr_2Si_2$ structure, and the lattice parameters of the samples were determined to be $a = b = 4.129 \pm 0.002$ Å, $c = 9.572 \pm 0.003$ Å. Within experimental errors, no changes of the lattice constant due to U substitution could be detected. In addition, susceptibility measurements have been used to check the magnetic properties. Their Neél temperatures were only slightly shifted as compared to those of undoped URu_2Si_2. A comprehensive study of the electrical and magnetic properties is given in [14].

We measured the Gd EPR with a Varian E-101 spectrometer at X-band frequency (9.3 GHz) using a 100 kHz field modulation. The spectrometer is equipped with an Oxford Instruments helium-flow cryostat. The samples were powdered and immersed in paraffin oil. Measurements were performed on grain-oriented samples in the temperature range 4.2 K $<$ T $<$ 290 K. The alignment of the grains was achieved with an 8 T superconducting magnet. The oriented samples allowed us to make measurements for a defined angle Θ between the crystallographic c-axis and the direction of the external magnetic field.

Figure 1 shows the absorption derivative of typical EPR spectra at 19 K with an external magnetic field parallel ($\Theta = 0^\circ$) and perpendicular ($\Theta = 90^\circ$) to the c-axis. The effect of grain alignment is clearly visible. The strongly exchange-narrowed [15] lineshape is Dysonian [16] with a dispersion fraction of almost 0 for all temperatures. Even at the lowest temperatures and the lowest Gd concentrations, the linewidth at resonance absorption was still exchange narrowed and individual lines which would be expected due to the crystal-field-split ground state collapse onto a single Lorentzian line. Crystal-field effects can only be detected via the angular dependence of the linewidth $\Delta H(\Theta)$ and the field at resonance absorption $H_{res}(\Theta)$ with respect to the external magnetic field.

The position of the resonance field H_{res} of the absorption line, which defines the g-value $g = h\nu/\mu_B H_{res}$, is shown in figure 2. For T $>$ 30 K the g-values, $g_\parallel$ and $g_\perp$, are both close to the free-electron value $g = 2$, and reveal a slight anisotropy only. For T $<$ 30 K and decreasing temperatures, $g_\parallel$ decreases significantly while $g_\perp$ remains almost constant, yielding a strong anisotropy below T_N. A similar behaviour has been detected in the ^{29}Si Knight shift in NMR experiments [8]. The strong decrease of $g_\parallel$ corresponds to the strong deviations from the static susceptibility $\chi_{0\parallel}$ for temperatures below 50 K [1] and can be attributed to the onset of strong spin correlations. We would like to point out that in normal magnetic compounds for temperatures above T_N, the g-values and line width increase significantly [17, 21], contrary to the findings for URu_2Si_2. In addition, in the AFM state usually an EPR signal cannot be observed [17, 21].

The temperature dependence of the linewidth $\Delta H(T)$ is shown in figure 3. With decreasing temperature $\Delta H(T)$ decreases almost linearly. For T $<$ 60 K the slope increases

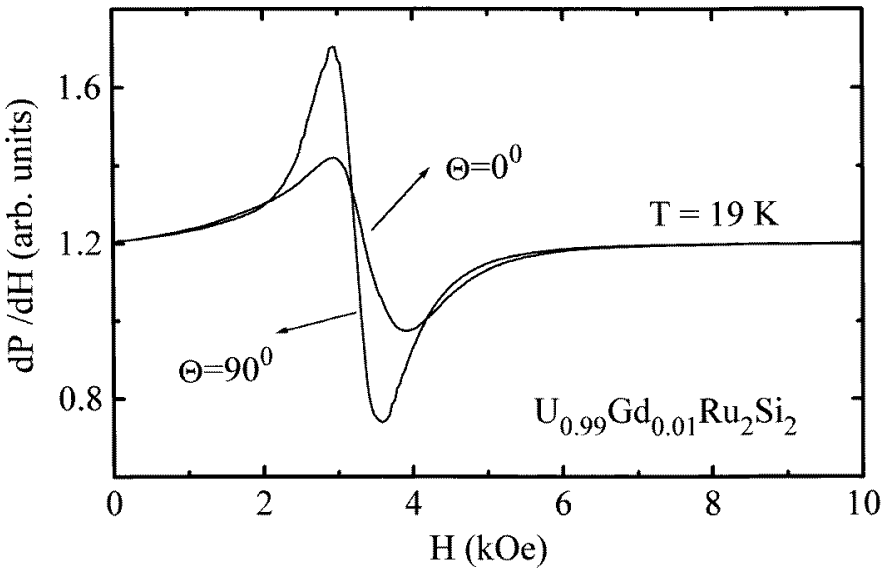

Figure 1. The absorption derivative dP/dH versus H for an oriented powder sample of URu_2Si_2:1% Gd. The observed anisotropy is due to the tetragonal crystal field.

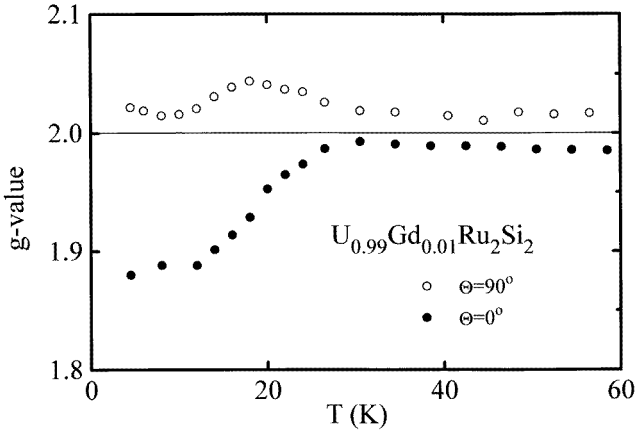

Figure 2. The position of the resonance line (g-value) for $\Theta = 0^\circ$ and $\Theta = 90^\circ$. The solid line represents the g-value of a free electron.

significantly and finally $\Delta H(T)$ decreases abruptly for $T < T_N$ (see the inset of figure 3). Note that the residual linewidth $\Delta H_0 = \Delta H(T = 0)$ is strongly anisotropic. This overall behaviour of $\Delta H(T)$ is very close to what has been observed in the temperature dependence of the spin–lattice relaxation rate $1/T_1$ in NMR experiments [7, 8, 9] and can easily be explained taking into account the two relaxation processes which are important in magnetic resonance experiments in heavy-fermion systems, namely [18]

$$\Delta H = \Delta H_0 + \Delta H_K + \Delta H_f. \tag{1}$$

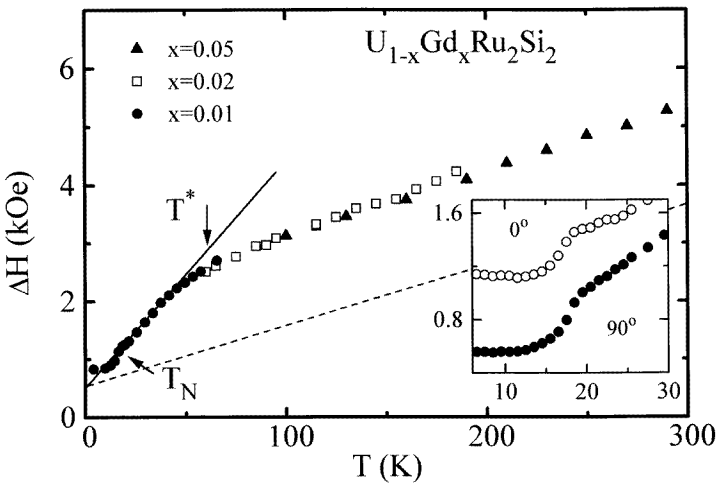

Figure 3. The temperature dependence of the linewidth ΔH versus T for grain-oriented polycrystalline URu_2Si_2, for different Gd concentrations. The inset shows ΔH in the vicinity of the AFM phase transition. The dashed line is a rough estimate of the Korringa behaviour that would be expected in a normal metal with unenhanced electronic masses.

Here ΔH_K is the Korringa relaxation which is determined by

$$\Delta H_K = bT. \tag{2}$$

The Korringa slope b depends on the square of the electronic density of states at the Fermi level. ΔH_f stems from the spin fluctuations of the 5f electrons which are transferred to the site of the Gd spin via the RKKY interaction [18, 20]

$$\Delta H_f \simeq T \sum_i K_i^2(\boldsymbol{r}_i)\chi_f''(\omega). \tag{3}$$

K_i denotes the effective coupling constant between the Gd moment and the f spin at site $\boldsymbol{r}_i$. $\chi_f''(\omega)$ is the dynamical susceptibility. The index f only refers to uranium 5f spins. Assuming a purely relaxational spin susceptibility and neglecting the summation over all neighbouring f ions, in the limit of low frequencies equation (3) can be simply rewritten as [18]

$$\Delta H_f \propto T\chi_0/\Gamma \tag{4}$$

where χ_0 is the static susceptibility and Γ is the magnetic relaxation rate which corresponds to the width of the quasielastic line in neutron scattering experiments [19]. Using this formalism, the temperature dependence of the width of the resonance absorption can easily be explained. For high temperatures ($T > 100$ K), we find

$$\Delta H = \Delta H_0 + b_0 T + C\sqrt{T}/(T - \Theta_{CW}). \tag{5}$$

Here we have assumed that the static susceptibility follows a Curie–Weiss law (this is true for $T > 130$ K, with a Curie–Weiss temperature $\Theta_{CW} = -65$ K [1]) and that Γ can be described by a square-root behaviour, as will be outlined below. b_0 represents the normal-metallic Korringa slope (the dashed line in figure 3). For low temperatures

$$\Delta H = \Delta H_0 + b_1 T \tag{6}$$

and b_1 is determined by the density of states in the heavy Fermi liquid (the solid line in figure 3). The temperature dependence of ΔH, as shown in figure 3, can easily be

described using a phenomenological model [18], or in the framework of a microscopic theory as recently demonstrated by Wright [22]. Figure 3 reveals that b_1 is enhanced only by a factor of 3. Comparing the Sommerfeld coefficient of URu_2Si_2 with that of a normal metal an enhancement of 10^4 would be expected if the heavy quasiparticles behave like true band states. Obviously the high electronic masses are almost localized close to the f site and the enormous difference in the observed relaxation behaviour is due to a strong dependence on the distance from the probe site to the f sites. This has been calculated in detail by Cox [20] who has shown that the mass-renormalization effects decay with the sixth power of the distance. In figure 3 the transition region between heavy-fermion behaviour and paramagnetic regime is close to 60 K (the arrow in figure 3), and indicates roughly the Kondo lattice temperature ($T^* \approx 70$ K), as determined from thermodynamic measurements [6].

Equations (1)–(3) enable us to derive the dynamic susceptibility, which can be written as follows:

$$\chi_f''(\omega_0) \propto (\Delta H - \Delta H_0 - bT)/T. \tag{7}$$

ω_0 is the frequency of the resonance absorption. Again we neglected all coupling constants. As the `normal'-metallic Korringa slope b is unknown because the relaxation channels cannot be separated, we used a value of 12 Oe K^{-1} (see the dashed line in figure 3), a value typically found in normal metals. This approach seems to be justified because the value of the Korringa slope has no significant impact on the following analysis.

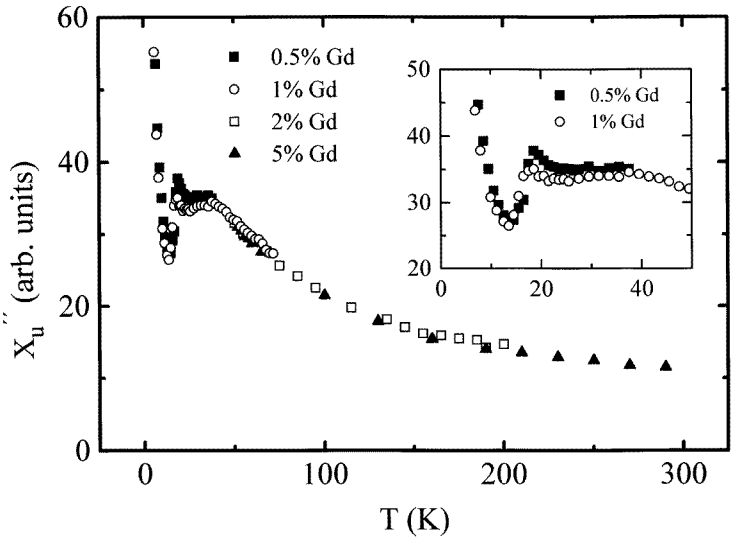

Figure 4. The dynamical susceptibility χ_U'' at the U site. Note (i) that for $T \approx 40$ K $\chi_U''(T)$ changes from a constant value to a Curie–Weiss-like behaviour and (ii) the anomaly at T_N.

The temperature dependence of the dynamical susceptibility $\chi_f''(\omega_0)$ is shown in figure 4. With decreasing temperatures, χ_f'' increases according to a Curie–Weiss law and becomes constant below 40 K. The AFM phase transition is indicated by a sharp anomaly in the dynamical susceptibility (see the inset of figure 4). It is interesting to note that in the dc susceptibility T_N can only be detected via the temperature derivative [1]. It is a clear and significant anomaly in $\chi_f''(T)$. The temperature dependence of the dynamical susceptibility

around T_N has also been determined from neutron scattering experiments [23] and a sharp peak has been detected at the magnetic phase transition temperature. Of course one has to keep in mind that neutron scattering and EPR experiments operate on vastly different time-scales.

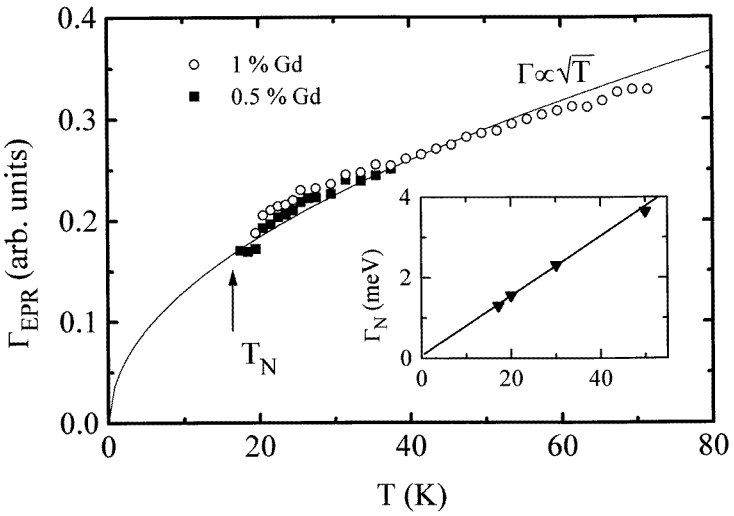

Figure 5. The quasielastic linewidth Γ_{EPR} as calculated from our EPR results. The solid line represents a square-root behaviour of $\Gamma_{EPR}(T)$. The inset shows the neutron scattering results $\Gamma_N(T)$ which reveal a linear temperature dependence in a limited temperature range (from [13]).

Finally we can use the experimentally determined static susceptibility to calculate the magnetic relaxation rate Γ_{EPR} using equation (4). The result is shown in figure 5 where Γ_{EPR} is plotted as a function of temperature. In the paramagnetic regime Γ_{EPR} follows a square-root dependence as has been calculated by Cox *et al* [19] for temperatures $T > T^*$. Our results are in good agreement with Γ_{NMR} as determined by Benakki *et al* [8] from the spin–lattice relaxation rate. But they are in clear contradiction to the neutron results. In these experiments [13] a linear temperature dependence of Γ_N has been detected in a limited temperature range.

3. Conclusions

We have reported measurements of the EPR linewidth and g-value of $U_{1-x}Gd_xRu_2Si_2$. For $T > 60$ K the relaxation of the Gd probe is predominantly due to its coupling to spin fluctuations. In the temperature range 20 K $< T < T^*$, ΔH reveals a linear, slightly enhanced Korringa slope indicating the presence of a heavy Fermi liquid. The enhancement is orders of magnitude less than expected from the bulk measurements. This result unambiguously demonstrates the rather localized character of the heavy quasiparticles. For $T < T_N$ the temperature dependence of the linewidth reveals the opening of a spin gap. The absence of any inhomogeneous broadening of the EPR absorption line in the AFM state can be explained by the extremely small ordered moments or via the band character of the magnetic order. The onset of heavy-fermion band magnetism is also indicated by the fact that magnetic order develops out of a purely heavy Fermi liquid, with fully compensated U moments. Finally we were able to derive the temperature dependence of the dynamical

susceptibility χ''_U at the U site and the quasielastic linewidth Γ_{EPR}. In the paramagnetic phase our data reveal that $\Gamma_{EPR}(T)$ follows a square-root behaviour as predicted by Cox *et al* [19], but our results are in disagreement with neutron scattering results, which show a linear T-dependence of Γ_N for $T > T_N$ [13].

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