

Paramagnetic resonance in GdRh_2Si_2

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Abstract. The paramagnetic properties of single-crystalline GdRh_2Si_2 are investigated by electron spin resonance (ESR) spectroscopy. The almost isotropic paramagnetic resonance is characterized by a spin dynamics of coupled Gd^{3+} – conduction electron spins. Towards magnetic ordering the spin relaxation is dominated by ferromagnetic in-plane spin fluctuations.

1. Introduction

GdRh_2Si_2 belongs to the rare-earth silicides with the tetragonal ThCr_2Si_2 structure which show exceptional magnetic properties, e.g. the antiferromagnetic (AFM) Kondo systems YbRh_2Si_2 [1] or CeRh_2Si_2 [2]. GdRh_2Si_2 possesses well localized magnetic moments with a weak magnetic anisotropy and displays AFM order below $T_N = 107$ K [3, 4]. It recently came into focus because of the spin-splitting of itinerant magnetic surface states induced by the ordering of the Gd 4f moments in the bulk [5].

Electron Spin Resonance (ESR) is a bulk sensitive method to investigate the spin dynamics of magnetic moments in metals where Gd^{3+} -spins are commonly used as probes [6]. This allows narrow lines because a direct spin relaxation to the lattice is not possible. The reason is the zero orbital magnetic moment of Gd^{3+} (pure spin ground state) and thus crystalline electric field effects as well as the magnetic anisotropy are weak. In this respect it is worth to note that in the heavy fermion metal YbRh_2Si_2 , despite a strong magnetic anisotropy, an ESR signal is observed as a consequence of the Kondo interaction [7].

In GdRh_2Si_2 well-localized Gd^{3+} magnetic moments serve as ideal probes for a spin resonance. Interestingly, the weak in-plane magnetic anisotropy [8] allows an observation of the in-plane resonance at standard microwave energies (X-band, 9.4 GHz) even in the magnetically ordered region. In this first report we focus on the Gd^{3+} ESR spectra in the paramagnetic region, which is easier to analyze and shall provide the basis for getting an understanding of the ESR signal in the AFM ordered regime.

2. Experiment and Results

For the ESR investigation we used high-quality single-crystalline GdRh_2Si_2 , the growth and characterization of which is described in Ref.[3]. We used a continuous-wave ESR spectrometer at X-band frequency ($\omega/2\pi = 9.40$ GHz) together with a nitrogen-flow cryostat allowing for temperatures between 100 and 300 K. Additional measurements at Q-band frequency ($\omega/2\pi = 34.07$ GHz) were done for $c \perp H$ (i.e. magnetic field is perpendicularly aligned to



the tetragonal c -axis) in the same temperature range by using a He-flow cryostat. In general, an ESR spectrometer allows to measure the absorbed power P of a transversal magnetic microwave field as a function of a static and external magnetic field $\mu_0 H$. A lock-in technique improves the signal-to-noise ratio by a field modulation which then yields the derivative of the resonance signal dP/dH as the measured quantity. The resulting ESR spectra were fitted with a Lorentzian function including the influence of the counter-rotating component of the linearly polarized microwave field [9]. From the fit we obtained the resonance field H_{res} and the linewidth ΔH (half-width at half maximum). In the paramagnetic region the resonance condition simply reads: $\omega/\gamma = H_{res}$ where $\gamma = g\mu_B/\hbar$ is the gyromagnetic ratio and g is the spectroscopic splitting factor (the “ESR g -factor”).

In the paramagnetic regime, i.e. for $T > T_N = 107$ K, the ESR spectra and their temperature dependence display a behavior as typically expected for well-defined local moments in a metallic environment and for temperatures nearby magnetic ordering [6, 10]. Fig. 1 shows $H_{res}(T)$ and $\Delta H(T)$ for two orientations of the external magnetic field relative to the tetragonal c -axis. An anisotropy within the tetragonal basal plane could not be detected above T_N . The spectra shapes are well defined Lorentzians (inset of Fig. 1). They are asymmetric due to a microwave penetration depth (≈ 0.003 mm) which is much smaller than the sample thickness (≈ 0.5 mm). The temperature dependences of H_{res} and ΔH result from the interaction between the magnetic moments of Gd^{3+} and conduction electrons. The anisotropy in the paramagnetic susceptibility $\chi \propto (T + \Theta)^{-1}$ leads to the observed weak temperature dependence of $H_{res}^{\parallel, \perp}$ in terms of the difference in the Weiss temperatures $\Theta_{\perp} - \Theta_{\parallel}$ [11]. The dashed grey lines in the upper frame of Fig. 1 fit the data with

$$g_{\parallel}^{\text{eff}} = g_{\parallel}^0 \left(1 + \frac{\Theta_{\perp} - \Theta_{\parallel}}{T + \Theta_{\parallel}} \right), \quad g_{\perp}^{\text{eff}} = g_{\perp}^0 \left(1 - \frac{0.5(\Theta_{\perp} - \Theta_{\parallel})}{T + \Theta_{\perp}} \right) \quad (1)$$

taken from Ref. [11] with $\Theta_{\parallel} = -6.5$ K and $\Theta_{\perp} = -9.2$ K which also consistently describe the paramagnetic susceptibility data (see Fig. 6 in [3]). [12] The g -factors, $g_{\parallel}^0 = 1.995$ and $g_{\perp}^0 = 2.001$, indicated in Fig. 1 are the high-temperature limits of Eqs. (1) and are consistent with the value reported for powdered samples $g = 1.995 \pm 0.01$ [13].

Towards high temperatures, the linewidth linearly depends on temperature and is determined by the relaxation of the coupled Gd^{3+} - conduction electron spin system towards the lattice (so-called “bottlenecked relaxation”) [13]. Approaching T_N , due to the decreasing fluctuations of the Gd^{3+} moments, the exchange-narrowing [14] gets suppressed. This leads to a critical divergence of ΔH^{crit} which is generally described by a power-law behavior (see p. 354 in [10]). We describe the total temperature dependence of ΔH in the paramagnetic regime as shown with the dashed line in Fig. 1 according to the following equation:

$$\Delta H = \Delta H^{\text{crit}} + b \cdot T + \Delta H_0 \quad \text{with} \quad \Delta H^{\text{crit}} = C_0 \left(\frac{T}{T_N} - 1 \right)^{-p}. \quad (2)$$

Here, C_0 and ΔH_0 are temperature independent parameters, $C_0^{\parallel} = 5.8$ mT, $C_0^{\perp} = 2.3$ mT and $\Delta H_0 = 5.5$ mT. The critical linewidth divergence is fitted with $p = 1$ and $T_N = 107$ K. $p = 1$ corresponds to a model for a 3D-Heisenberg ferromagnet whereas a 3D-Heisenberg antiferromagnet would correspond to $p = 1.7$ (see p. 358 in [10]). Therefore, ferromagnetic fluctuations in the plane seem to dominate the spin dynamics in agreement with in-plane ferromagnetically ordered layers but in contrast to their AFM staggered order [8]. Although ΔH^{crit} describes the data within experimental error reasonably well it cannot account for the fact that the maximum of the observed linewidth occurs a few degrees above T_N . This effect is observed also in other ferromagnets and is related to the field-dependence of the linewidth-divergence, see Ref. [15], for instance. The linear parts fit to the slopes $b_{\perp} = 0.18$ mT/K and

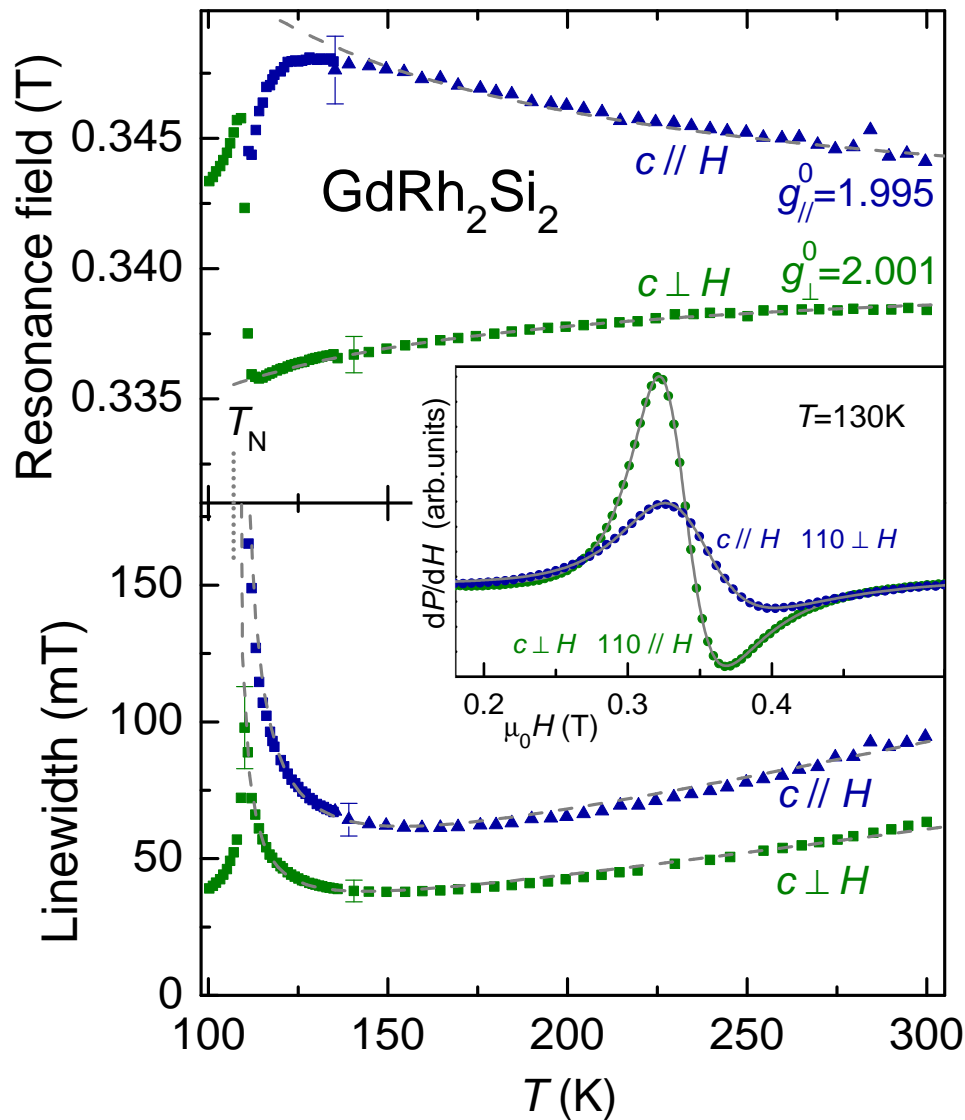


Figure 1. (Color online) Temperature dependence of the resonance field H_{res} and linewidth ΔH as obtained from fitting the X-band (9.4 GHz) spectra with Lorentzian shapes, see inset, solid lines. The dashed lines describe $H_{res}(T)$ with a weak anisotropy in the paramagnetic susceptibility (parameters see Eqs. 1 and main text) with high- T limits of the ionic g -values as indicated. The $\Delta H(T)$ data could be described by dashed lines according Eq.(2) with a dominant critical behavior for temperatures approaching $T_N = 107$ K and with a linear dependence at elevated temperatures (parameters see main text).

$b_{\parallel} = 0.28$ mT/K, again in agreement with data of powdered samples [13]. As compared to the X-band data the paramagnetic linewidth of the Q-band data (measured for $c \perp H$ only) could be described with the same parameters of the critical part but with clearly different ones in the linear part: $b_{\perp}^Q = 0.07$ mT/K and $\Delta H_0^Q = 24$ mT.

3. Discussion and Conclusion

We have shown that at comparatively small energies (X-band, 9.4 GHz \equiv 0.04 meV and Q-band, 34 GHz \equiv 0.14 meV) the internal magnetic fields and the spin dynamics of GdRh₂Si₂ can be investigated by electron spin resonance of the Gd³⁺ spins. In the paramagnetic regime, $T > T_N = 107$ K, a common Gd³⁺ collective-mode resonance is observed. The ESR g -values follow nicely a temperature dependence as expected for anisotropic exchange-coupled paramagnets [11]. The spin dynamics is characterized by a coupled Gd³⁺ - conduction electron spin system and a linewidth divergence towards T_N determined by a slowing down of in-plane ferromagnetic fluctuations.

References

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