High pressure μSR studies on single crystalline gadolinium

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Muon spin rotation data on a single crystalline gadolinium sample have been obtained as function of temperature and hydrostatic external pressure up to 0.6 GPa.

In the ferromagnetic state the application of pressure has a strong influence on the whole spin-turning-process of the spontaneous magnetization: The onset of the spin-turning is shifted towards lower temperatures with a rate of approximately $\mathrm{d}T_{\mathrm{st}}/\mathrm{d}p \approx -50$ K/GPa. Higher values of the turning angle up to $\vartheta_{\mathrm{ext}} = 90^\circ$ can be reached and stabilized over a wider temperature range. At low temperatures the magnetization turns back again.

In the paramagnetic regime the data show a similar behaviour as under ambient pressure: both the Knight-shift and the muon spin relaxation show deviations from their high temperature behaviour in the critical regime below $(T - T_{\mathrm{C}}) < 10$ K. So there is no indication of pressure induced effects in this temperature range.

1. Introduction

Gadolinium metal crystallizes in the double hexagonal close packed structure like all heavy rare earth elements. It is the only elemental rare earth which shows a direct transition from paramagnetic to simple ferromagnetic order. The Curie temperature is close to room temperature ($T_{\mathrm{C}} = 292.8$ K).

The ferromagnetic state of gadolinium is characterized by a temperature dependent rotation of the direction of easy magnetization away from its orientation at T_{C} being parallel to the hexagonal c-axis. This can be seen in neutron- [1] and torque- [2] as well as in μSR-data [3,4].

The muon comes to rest at the octahedral interstitial site. Here it is sensitive not only to the isotropic contact field (originated in the spin polarization of the conduction electrons) but also to the anisotropic dipolar field of the localized Gd^{3+} moments

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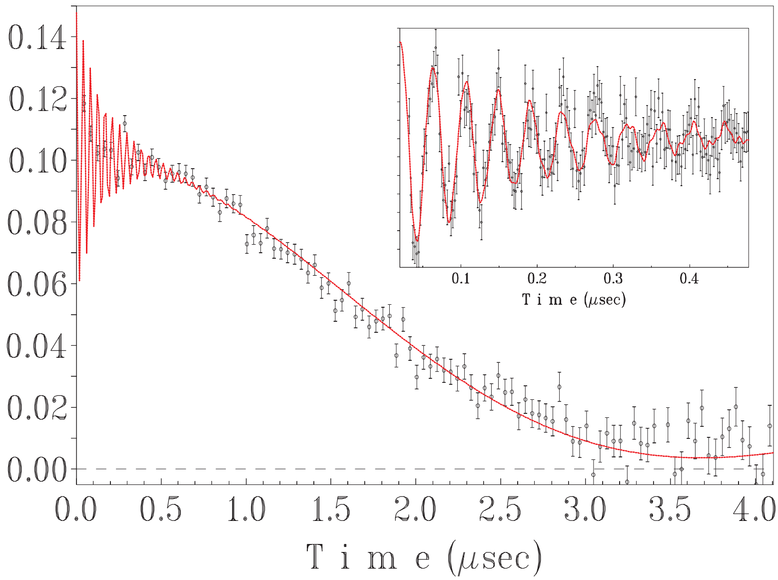


Fig. 1. μSR-asymmetry-spectrum of ferromagnetic gadolinium inside a high pressure cell made of CuBe. Note the small amplitude and the high damping rate of the spontaneous rotation signal lying on the zero field spectrum of the pressure cell.

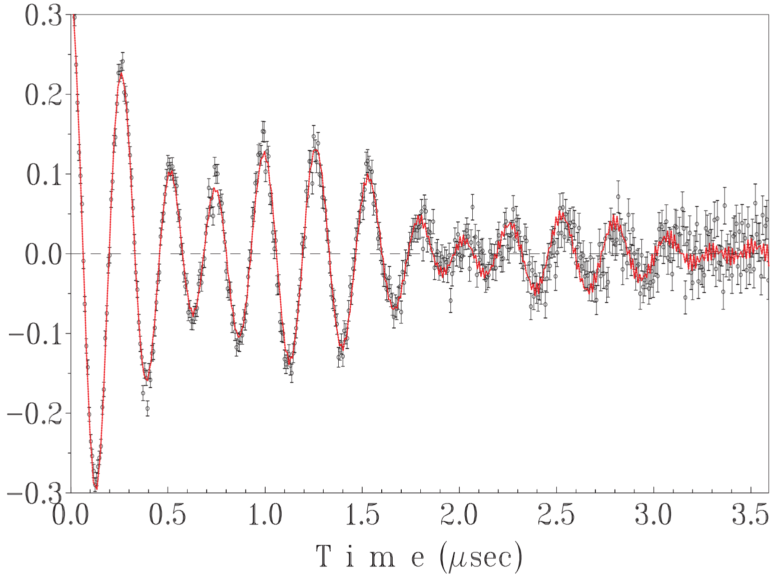


Fig. 2. μSR-asymmetry-spectrum of paramagnetic gadolinium inside a high pressure cell made of CuBe. Note the beating of two rotation signals, originated in the local field B_{μ} inside the sample and the external field B_{ext} inside the pressure cell.

surrounding the muon. Therefore it is an excellent tool to study the changes in the spin arrangement.

In the paramagnetic regime μSR provides two observables, the paramagnetic frequency- or Knight-shift and the relaxation rate, to study the static as well as the dynamic behaviour of magnetic short range order. Previous μSR measurements in this temperature range observed the transition from isotropic to dipolar critical behaviour below $T - T_C \approx 10$ K and the existence and increase of magnetic clusters in this critical regime [5].

2. Experimental details

The described μSR measurements were performed at the high-pressure-μSR-spectrometer at PSI, Switzerland, in a decay muon beam. A single crystalline rod of gadolinium with 7 mm in diameter and a length of 40 mm was mounted inside a high pressure cell made of CuBe, with the crystal c-axis perpendicular to the initial muon spin polarization. Spectrometer and cell are described elsewhere [6]. The data were obtained in the temperature range 15 K $\leqslant T \leqslant$ 340 K and under hydrostatic external pressures up to $p = 0.6$ GPa. The measurements of the spontaneous muon spin frequency were done in zero applied field, the Knight-shift data in a transverse field of $B_{ext} = 28$ mT. The data were analyzed with two signals, originating in the sample and the high pressure cell with a signal ratio of 1:2. Figures 1 and 2 show typical μSR spectra obtained in the ferro- and paramagnetic range.

3. Results and discussion

3.1. Ferromagnetic range

In the ferromagnetic ordered state of gadolinium one can observe a spontaneous muon spin rotation frequency $\omega_\mu = \gamma_\mu B_\mu$, which is directly connected to the local magnetic field B_μ at the muon site (fig. 1).

The temperature dependence of B_μ shows significant deviations from the magnetization curve as seen in fig. 3. This can be explained by describing B_μ as the vector sum of the isotropic Fermi contact field B_{fc} (roughly proportional to the magnitude of magnetization) and of the anisotropic dipolar field B_{dip} (strongly dependent on the direction of magnetization), which is able to extract the turning angle ϑ between the ferromagnetic easy axis and the hexagonal c-axis [3].

The data measured under ambient pressure with our high-pressure-spectrometer are in accordance with previous μSR results [3,4] and reflect the well-known behaviour of the spin-turning-process (fig. 3(a)).

Just below the magnetic transition $T_{st} \leqslant T < T_C = 292.8$ K, the magnetization is aligned parallel to the hexagonal c-axis. At $T_{st} \approx 230$ K, the so-called spin-turning-

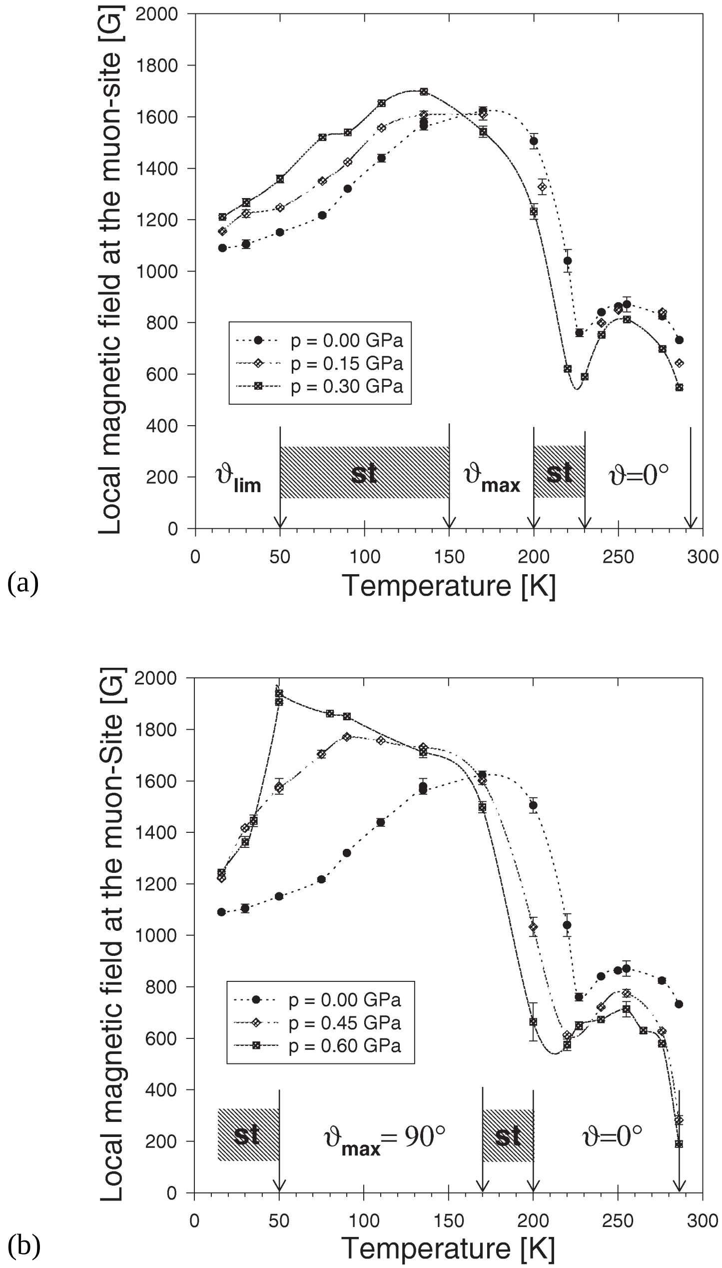


Fig. 3. Temperature dependence of the local magnetic field $B_{\mu}(T)$ at the muon site in ferromagnetic gadolinium under external pressure up to $p \leqslant 0.6\,\text{GPa}$. The marked regions characterize the spin-turning-process under ambient pressure (a) and under the maximum applied pressure $p = 0.6\,\text{GPa}$ (b) as described in the text.

temperature, it starts to turn away from this direction. This is reflected by the strong increase of B_μ in the temperature range $200\,\mathrm{K} \leqslant T \leqslant T_{\mathrm{st}}$. At $T \approx 200$ K a maximum turning angle $\vartheta^{0.0}_{\mathrm{max}} \approx 60^\circ$ is reached and stays down to $T \approx 150$ K. By lowering the temperature further, the easy axis slowly turns back, which causes the smooth decrease of the local field B_μ. At $T \approx 50$ K a low temperature value of $\vartheta^{0.0}_{\mathrm{lim}} \approx 30^\circ$ is reached.

As one can see in fig. 3(b), the application of pressure has significant influence to the spin-turning-process described above.

Above the spin-turning-temperature $T_{\mathrm{st}} \leqslant T \leqslant T_{\mathrm{C}}$, where the ferromagnetic easy axis is still parallel to the hexagonal c-axis, a decrease of the local field with pressure $\mathrm{d}B_\mu/\mathrm{d}p < 0$ is seen. The values of B_μ under pressure are lower than one would expect if only considering the pressure induced shift of the ordering temperature ($\mathrm{d}T_{\mathrm{C}}/\mathrm{d}p = -14.0(2)$ K/GPa [7]). We interpret this as an effect of an increased spin polarization of the conduction electrons with reduced volume which leads to an enhanced contribution of the negative contact field $\mathrm{d}\ln B_{\mathrm{fc}}/\mathrm{d}p > 0$ under pressure.

The onset of the spin-turning is shifted towards lower temperatures with a rate of approximately $\mathrm{d}T_{\mathrm{st}}/\mathrm{d}p \approx -50$ K/GPa in accordance with the results of macroscopic measurements [8,9], whereas the rate of the initial spin-turning, i.e., $\mathrm{d}B_\mu(T)/\mathrm{d}T$ is nearly pressure-independent.

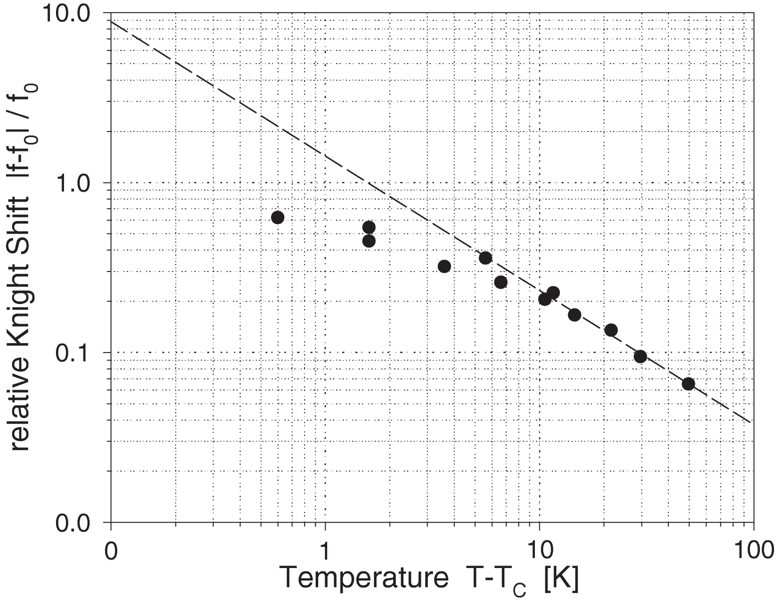
Applying the procedure of Denison et al. [3] for extracting the turning angle ϑ from these μSR data under an external pressure of $p_{\mathrm{ext}} = 0.6$ GPa (with adequate corrections of the crystal structure and the magnetization), a maximum value of $\vartheta^{0.6}_{\mathrm{max}} \approx 90^\circ$ is reached at $T = 170$ K. The magnetization persists in this perpendicular position down to $T = 50$ K. In contrast to the situation under ambient pressure, one now observes a continuous increase of B_μ in the temperature regime $50\ \mathrm{K} \leqslant T \leqslant 170$ K, which is proportional to the temperature dependence of the spontaneous magnetization. At $T = 50$ K, the easy-axis turns back again as seen in the rapid decrease of the local field B_μ.

3.2. Paramagnetic range

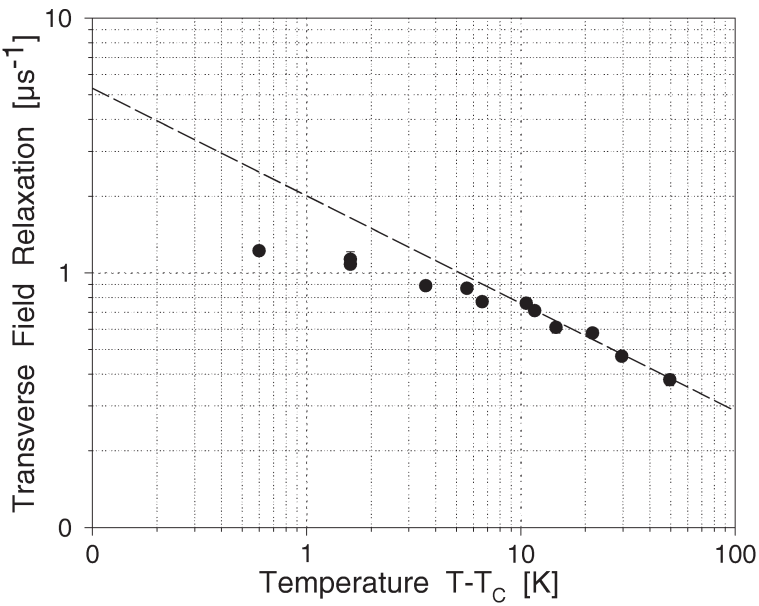
In the paramagnetic state we have measured the paramagnetic frequency- or Knight-shift $\Delta f/f_0 \equiv (B_\mu - B_{\mathrm{ext}})/B_{\mathrm{ext}}$, which is proportional to the local magnetic susceptibility $\chi_l \propto |T - T_{\mathrm{C}}|^\gamma$ and the muon spin relaxation rate λ_μ proportional to the correlation time $\tau_{\mathrm{c}} \propto |T - T_{\mathrm{C}}|^w$ of the localized magnetic moments surrounding the muon. Due to experimental restrictions (high-pressure-cell) we had to use a nonspherical single-crystalline sample. Therefore our results can only be qualitatively compared to the previous μSR measurements done on a polycrystalline gadolinium-sphere [5].

Figure 4 shows the temperature dependence of the relative Knight-shift (a) and the muon relaxation rate (b) in a double logarithmic scale.

As under ambient pressure, both the Knight-shift $|\Delta f/f_0|$ and the muon spin relaxation λ_μ deviate from their paramagnetic behaviour in the critical regime below $T - T_{\mathrm{C}} \approx 10$ K. The reduced exponent $\gamma < 1$ of the local susceptibility χ_l is in contrast to the critical behaviour of the macroscopic susceptibility ($\gamma = 1.23(2)$ [10]) and



(a)



(b)

Fig. 4. Temperature dependence of the relative Knight-shift $|\Delta f/f_0|$ (a) and of the muon relaxation rate λ_μ (b) in paramagnetic gadolinium, measured in an applied transverse field of $B_{\text{ext}} = 28$ mT and under an external pressure of $p_{\text{ext}} = 0.6$ GPa and plotted in a double-logarithmic scale.

has to be explained by the onset of local magnetic order (clusters) around the muon, which counteracts the free paramagnetic Knight-shift. The reduction of the dynamic exponent w reflects the cross-over from isotropic to dipolar critical behaviour [5].

In contrast to the measurements under ambient pressure we cannot observe a pure Curie–Weiss behaviour ($\gamma = 1$) up to the highest temperature measured $T - T_C = 50$ K. This has to be checked by further measurements over a wider temperature range.

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