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High-pressure μSR studies on elemental rare earth metals

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Abstract

Muon-spin rotation data on single-crystalline samples of the heavy rare-earth metals Dy and Ho in their antiferromagnetic and ferromagnetic temperature regimes ($20\ \mathrm{K} < T < T_N$) have been obtained as function of hydrostatic external pressure up to 0.9 GPa using the He-gas high-pressure system installed at the decay muon beam μE1 at PSI. Due to the absence of pressure-dependent changes of the moment orientation (spin-turning) as observed in previous high-pressure measurement on ferromagnetic Gd, the pressure coefficients $(\partial B_\mu/\partial p)_T$ in Dy and Ho are comparatively small, but nevertheless temperature-dependent. The volume dependence of the contact field is extracted. © 2000 Elsevier Science B.V. All rights reserved.

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The μSR data on the helical antiferromagnets Dy and Ho under the application of hydrostatic external pressure up to $p_{\max} \leqslant 0.9$ GPa presented in this study are an addition to our measurements under ambient pressure on these metals also presented at this conference [1]. They also are a continuation of our previous high-pressure μSR experiments on ferromagnetic Gd [2]. The latter work has a strong effect of pressure (or reduced volume) on the spin turning process $\theta(T;p)$ of the ferromagnetic moments away from the c-axis. This effect can be observed well by μSR spectroscopy via characteristic changes with temperature and pressure of the local magnetic field B_μ at the interstitial muon site $\boldsymbol{R}_\mu$. The situation at ambient pressure has been discussed in detail by Graf et al. [3]. In the vector sum

$$\boldsymbol{B}_\mu(T;p) = \boldsymbol{B}_{\mathrm{fc}}(T;p) + \boldsymbol{B}_{\mathrm{dip}}(\theta(T;p), \boldsymbol{R}_\mu),$$

the strong anisotropy of the dipolar field causes quite a different temperature dependence of $\boldsymbol{B}_{\mathrm{dip}}(\theta(T;p))$ than the Brillouin-like dependence of the isotropic contact field $B_{\mathrm{fc}}(T;p)$ generated by the polarization of conduction electrons. An appropriate tensor formalism allowed the extraction of $\theta(T;p)$. The effect is very pronounced in Gd since B_{dip} and B_{fc} are of comparable magnitude. In contrast to Gd, the strong magnetic anisotropy acting in Dy and Ho confines the magnetic moments to the basal plane ($\theta = 90°$) over the whole temperature regime $20\ \mathrm{K} < T < T_N(p)$ investigated. No

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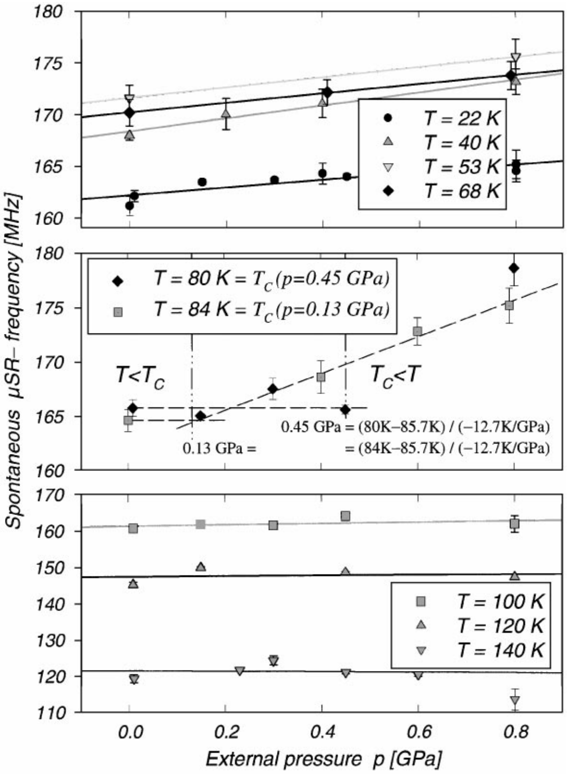

Fig. 1. Pressure dependence of the spontaneous muon precession frequency $v_\mu(p)$ in the ferromagnetic and helical antiferromagnetic temperature regime of single-crystalline Dysprosium. The increase of the muon precession frequency $v_\mu(p)_T$ under pressure at the temperatures $T = 80$ and 84 K (middle) reflects the pressure induced shift $\partial T_C/\partial p = -12.7$ K/GPa [5] of the ferromagnetic ordering temperature and is consistent with a value $T_C(p = 0) \approx 85.7$ K under ambient pressure.

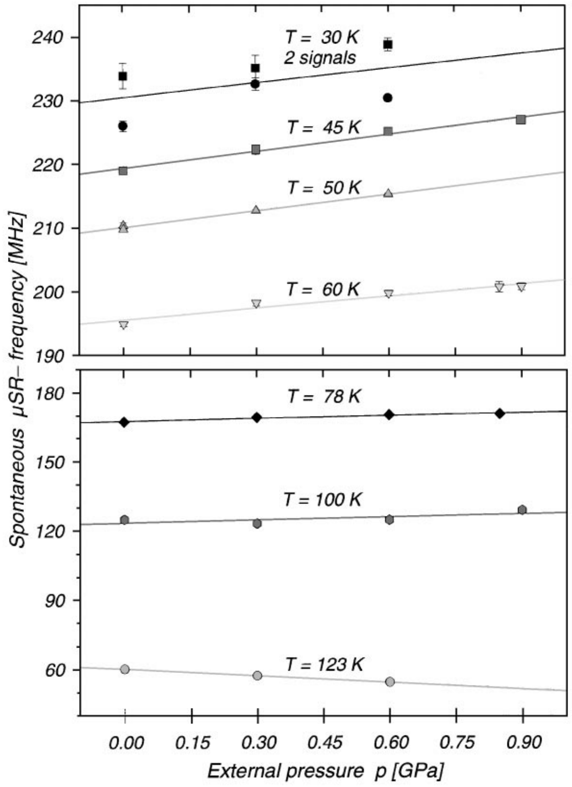

Fig. 2. Pressure dependence of the spontaneous muon precession frequency $v_\mu(p)$ in the helical antiferromagnetic temperature regime of single-crystalline Holmium.

indication of changes in spin orientation are seen in these metals, as expected. Therefore, we focus our attention on the pressure dependence of the conduction electron polarization mediated by the contact field $B_{fc}(T; p)$.

All measurements were performed at PSI, Switzerland, using the decay muon beamline μE1 in conjunction with the He-gas high-pressure system described elsewhere [4]. The samples consisted of three single-crystal rods each with a diameter of 7 mm and a total length of 20 mm. They were mounted in the central bore of a cylindrically symmetrical high-pressure cell made of CuBe. They were oriented with their c-axis parallel to the initial muon-spin polarization $\boldsymbol{P}_\mu$. This leads to a maximal amplitude of the observed rotation signal.

The antiferromagnetic structure of Dy and Ho below $T_N(p = 0) \approx 180$ and 131 K, respectively, is an helix with the moments confined to the basal plane, the helical-axis being the crystalline c-axis. Below $T_C(p)$ the moments order ferromagnetically along the a-axis in Dy, whereas in Ho the spiral ordering of the basal plane components persists, which, in combination of a ferromagnetic ordered axial moment, leads to a shallow conical ferromagnetic spin structure. The temperature limitations of the closed-cycled-refrigerator used for cooling the high-pressure cell restricted the measurements on Ho to the antiferromagnetic regime $T_C(p) \leqslant 20\text{ K} < T < T_N(p)$. In Dy with a Curie temperature of $T_C(p = 0) \approx 86$ K both ordered regimes could be studied. Detailed information about the magnetic structures and the temperature

Table 1

Pressure coefficients $(\partial B_\mu/\partial p)_T$ of the measured local magnetic field $B_\mu(T)$ at various temperatures as derived by a linear regression fit to the spontaneous μSR frequency data in the ordered regime of single-crystalline Dy and Ho as seen in Figs. 1 and 2, respectively. The errors refer only to the linear regression fit. The dipolar field $B_{\text{dip}}(T)$ and its pressure derivatives $(\partial B_{\text{dip}}/\partial p)_T$ are calculated under consideration of the pressure-dependent negative shift of the ordering temperature $(\partial T_{\text{ord}}/\partial p) < 0$, leading to a reduced magnetic moment $(\partial \ln \mu_S/\partial p)_T < 0$ and the compressibility $(-\partial \ln V/\partial p)_T > 0$, i.e. the reduction of the volume under pressure. The combination of these measured and calculated values gives the contact field $B_{\text{fc}}(T) = B_\mu(T) - B_{\text{dip}}(T)$ of the polarized conduction electrons at the interstitial site and its pressure coefficient $(\partial B_{\text{fc}}/\partial p)_T$

Dysprosium		FM		AFM		
Temp	(K)	22	68	100	120	140
$B_\mu^{\text{regr}}(T)$	(kG)	+ 11.97(3)	+ 12.56(1)	+ 11.91(7)	+ 10.9(1)	+ 9.1(2)
$(\partial B_\mu/\partial p)_T$	(kG/GPa)	+ 0.27(6)	+ 0.33(1)	+ 0.13(15)	+ 0.06(29)	− 0.06(4)
$B_{\text{dip,octa}}(T)$	(kG)	+ 13.12	+ 12.37	+ 11.09	+ 10.12	+ 8.87
$(\partial B_{\text{dip}}/\partial p)_T$	(kG/GPa)	+ 0.31	+ 0.28	+ 0.19	+ 0.09	− 0.02
$B_{\text{fc,octa}}(T)$	(kG)	− 1.15(3)	+ 0.19(1)	+ 0.82(7)	+ 0.76(14)	+ 0.21(18)
$(\partial B_{\text{fc}}/\partial p)_T$	(kG/GPa)	− 0.04(6)	+ 0.06(1)	− 0.06(15)	− 0.03(29)	− 0.04(4)
Holmium		AFM				
Temp	(K)	44	50	66	78	100
$B_\mu^{\text{regr}}(T)$	(kG)	+ 16.18(3)	+ 15.50(2)	+ 14.43(4)	+ 12.36(3)	+ 9.10(13)
$(\partial B_\mu/\partial p)_T$	(kG/GPa)	+ 0.65(6)	+ 0.64(4)	+ 0.46(7)	+ 0.31(5)	+ 0.33(23)
$B_{\text{dip,octa}}(T)$	(kG)	+ 11.44	+ 11.17	+ 10.31	9.51	7.55
$(\partial B_{\text{dip}}/\partial p)_T$	(kG/GPa)	+ 0.21	+ 0.18	+ 0.10	+ 0.01	− 0.20
$B_{\text{fc}}(T)$	(kG)	+ 4.74(3)	+ 4.33(2)	+ 4.13(4)	+ 2.85(3)	+ 1.55(13)
$(\partial B_{\text{fc}}/\partial p)_T$	(kG/GPa)	+ 0.45(6)	+ 0.45(4)	+ 0.36(7)	+ 0.30(5)	+ 0.54(23)

dependence of the local magnetic field $B_\mu(T; p = 0)$ at ambient pressure can be found in Ref. [1].

Figs. 1 and 2 show the spontaneous muon precession frequency $\nu_\mu(p)_T = (\gamma_\mu/2\pi)B_\mu(p)_T$ as a function of pressure at various temperatures for Dy and Ho, respectively. The linear regression fit leads to the pressure coefficients $(\partial B_\mu/\partial p)_T$ of the local magnetic field $B_\mu(T)$ at the muon site. They are summarized in Table 1 together with the calculated pressure coefficients $(\partial B_{\text{dip}}/\partial p)_T$ of the dipolar field $B_{\text{dip}}(T)$.

Just below the antiferromagnetic transition the main influence of pressure on the dipolar field $B_{\text{dip}}(p)_T$ comes from the reduction of the magnetic moment $(\partial \ln \mu/\partial p)_T$. This in turn is predominantly due to the negative shift with pressure of the ordering temperature $\partial T_N/\partial p \approx -4.1(1)$ K/GPa for Dy and $\partial T_N/\partial p \approx -4.8$ K/GPa for Ho [5]. The change of moment is then approximated by an appropriate adjustment of the normalized magnetization curve to the change of magnetic ordering temperature. At lower temperatures, that is below $(T/T_N)_p < 0.75$ (Dy) or < 0.6 (Ho), the change of dipolar field $(\partial B_{\text{dip}}/\partial p)_T$ arises mainly from an increase of the bulk magnetization as a consequence of reduced volume $(-\partial \ln V/\partial p)_T$. This leads to a positive pressure coefficient $(\partial B_\mu/\partial p)_T > 0$ of the measured local magnetic field as can be seen in Figs. 1 and 2. Values of the isothermal compressibility $(-\partial \ln V/\partial p)_T$ were derived from the temperature dependence of the elastic constants [6].

As stated above, the magnetic moments have no axial component in the temperature ranges covered by this study and the angular dependence of the dipolar field $\boldsymbol{B}_{\text{dip}}$ need not to be considered. The application of external pressure then (in contrast to the case of Gd) changes only the magnitude of the local magnetic field $B_\mu(p)_T$, but not the basic Brillouin-like shape of its temperature dependence $B_\mu(T)_p$. The dipolar field $\boldsymbol{B}_{\text{dip}}$ in Dy and Ho is nearly fully aligned with the Fermi contact field $\boldsymbol{B}_{\text{fc}}$ and thus $B_{\text{fc}}(T)_{p=0}$ and its pressure coefficient $(\partial B_{\text{fc}}/\partial p)_T$ can be extracted with ease from the combination of the measured and calculated values $B_\mu(T)_{p=0}$ and $B_{\mathrm{dip}}(T)_{p=0}$ or $(\partial B_\mu/\partial p)_T$ and $(\partial B_{\mathrm{dip}}/\partial p)_T$, respectively. Table 1 gives a summary.

The contact field $B_{\mathrm{fc}}(T)$ in Dy is the relatively small difference between the two fields $B_\mu(T)_p \approx B_{\mathrm{dip}}(T)_p$ which are of comparable magnitude. In addition, its pressure coefficient nearly vanishes ($(\partial B_{\mathrm{fc}}/\partial p)_T \approx 0$). The uncertainties in both the contact field and its pressure coefficient, which cannot easily be reduced further, do not allow to draw clear conclusions about the volume dependence of the conduction electron polarization from the existing data.

The pressure-induced shift of the ferromagnetic ordering temperature $T_C(p)$, indicated by the small discontinuity or local minimum of the magnetic field $B_\mu(T_C)$ (see Ref. [1]), can be observed by the increase of the muon precession frequency $\nu_\mu(p)_T$ under pressure at the temperatures $T = 80$ and 84 K in Fig. 1 (middle). Under the assumption of a pressure shift of $\partial T_C/\partial p = -12.7$ K/GPa [5] the experimental data in the vicinity of the ferromagnetic transition lead to an ordering temperature of $T_C(p=0) \approx 85.7$ K under ambient pressure.

The values of the contact field $B_{\mathrm{fc}}(T)_p$ and its pressure coefficient $(\partial B_{\mathrm{fc}}/\partial p)_T$ in the antiferromagnetic range of Ho are one order of magnitude larger than in Dy (see Table 1) and lead to a more systematic relative pressure or volume dependence in the order of $(\partial \ln B_{\mathrm{fc}}/\partial p)_T \approx 0.4$ kG/GPa or $(\partial \ln B_{\mathrm{fc}}/\partial \ln V)_T \approx -3.7$. Also visible in Fig. 2 is the existence of two spontaneous muon frequencies at $T = 30$ K just above the ferromagnetic transition as an indication for various magnetic environments of the muon probe as consequence of a 'spin–slip structure' (see Ref. [1]), with both frequencies depending on pressure.

For the time being, these results on the pressure dependence of $B_{\mathrm{fc}}(p)_T$ must stand as they are at this stage. Theoretical calculations are to our knowledge not available. They need to be carried out with the presence of the muon taken into account, since the positive charge of the muon enhances the local spin density of the conduction electrons.

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References

[1] E. Schreier, M. Ekström, O. Hartmann, G.M. Kalvius, R. Wäppling, A. Marelius, S. Henneberger, F.J. Burghart, A. Kratzer, Physica B 289–290 (2000), These Proceedings.

[2] E. Schreier, S. Henneberger, F.J. Burghart, A. Kratzer, G.M. Kalvius, O. Hartmann, M. Ekström, R. Wäppling, Hyperfine Interactions 104 (1997) 311.

[3] H. Graf, W. Hofmann, W. Kündig, P.F. Meier, B.D. Patterson, W. Reichart, Solid State Commun. 23 (1977) 653.

[4] K. Mutzbauer, O. Hartmann, A. Kratzer, S. Henneberger, H.-H. Klauß, F.J. Litterst, R. Wäppling, G.M. Kalvius, Physica B 190 (1993) 40.

[5] H. Bartholin, D. Bloch, J. Phys. Chem. Solids 29 (1968) 1063.

[6] S.B. Palmer, E.W. Lee, Proc. Roy. Soc. A 327 (1972) 519.