



Internal fields in magnetically ordered dysprosium, holmium and erbium

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Abstract

Muon spin rotation data on single-crystalline samples of the heavy rare earth metals Dy, Ho and Er have been obtained as function of temperature in both the antiferromagnetic (afm) and the ferromagnetic (fm) state. In the afm state the temperature dependence of the spontaneous muon spin precession frequency consistently exhibits Brillouin-like behavior. In the fm state we observe, in all three metals, a decrease of frequency on cooling, while one expects a nearly temperature-independent saturation ($T \rightarrow 0$) behavior. Although the origin of this feature is not clear, it definitely cannot be connected to a spin reorientation. It is suggested that spontaneous bulk magnetization caused by the strong magnetic anisotropy might be responsible. In Dy and Ho only minor irregularities are seen at T_C . In contrast, Er shows a huge drop of the spontaneous frequency at the fm transition temperature, which can be directly traced to the behavior of the dipolar field component at the muon site. Saturation values for the dipolar and the contact field at the muon site for the three metals are given. © 2000 Elsevier Science B.V. All rights reserved.

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The spontaneous muon precession frequency ν_μ observed in the ordered state of magnetic materials is proportional to the local magnetic field \vec{B}_μ at the interstitial muon site \mathbf{R}_μ

$$\nu_\mu = (\gamma_\mu/2\pi) \cdot \vec{B}_\mu(\mathbf{R}_\mu)$$

with $(\gamma_\mu/2\pi) = 135.5342$ MHz/T being the gyromagnetic ratio of the muon. In the absence of an external magnetic field this local magnetic field $\vec{B}_\mu(\mathbf{R}_\mu)$ sensed by the muon is composed of the

vector sum of two magnetic field contributions having different origins but comparable orders of magnitude

$$\vec{B}_\mu(T) = \vec{B}_{fc}(T) + \vec{B}_{dip}(T).$$

The Fermi contact field $B_{fc}(T)$ is due to the spin-polarized conduction electrons. The dipolar field $B_{dip}(T)$ is directly generated by the magnetic moments on the surrounding ions. The combination of the measured local magnetic field $B_\mu(T)$ with the calculated dipolar field $B_{dip}(T)$ at the muon site allows both a local test of the magnetic structures (as proposed by neutron scattering data) and a determination of the interstitial contact field $B_{fc}(T)$ which is difficult to treat theoretically.

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In Dy the magnetic moments are confined to the basal plane. Between $T_N \approx 180$ K and $T_C \approx 86$ K, a helical antiferromagnetic spin structure is formed. The helix angle decreases with reduced temperature. At T_C , an orthorhombic lattice distortion occurs and all spins are ferromagnetically aligned along the orthorhombic a -axis [1].

In the helical spin structure of Ho, formed below $T_N \approx 131$ K, the hexagonal anisotropy leads to distortions of the regular helix at commensurability points with the crystallographic lattice, which produces the so-called ‘spin-slip-structures’. Below $T_C \approx 20$ K, a weak ferromagnetic moment along the c -axis develops. The spiral order of the basal plane components is still present, resulting in a shallow conical ferromagnetic structure with a cone angle of $80.5^\circ \leq \theta \leq 90^\circ$. The hexagonal anisotropy leads to a bunching of the moments along the b -axis instead of a regular helix with $\langle \phi \rangle = 30^\circ$ [1].

Erbium shows two afm regimes: Below $T_{N,\parallel} \approx 85$ K a sinusoidal c -axis modulation (CAM) of the axial moment is present. At $T_{N,\perp} \approx 53$ K an additional helical ordering of the basal plane components takes place. Higher-order harmonics of both modulations lead to a so-called ‘anti-phase-domain’ (APD) structure. Several ‘spin-slip-transitions’ occur here as well. The fm structure below $T_C \approx 20$ K is conical with a cone angle of $\theta \leq 29^\circ$ [1].

The μ SR experiments were carried out at the decay muon beamlines μ E1 and μ E4 of PSI, Switzerland. Data were obtained in zero applied field from 10 to 200 K in a closed-cycle refrigerator and a ^4He -cryostat and below 10 K in a ^3He -cryostat. The single-crystal rods (7 mm diameter \times 20 mm length) were orientated with the initial muon spin polarization parallel to the crystal c -axis ($\mathbf{P}_\mu \parallel \mathbf{c}$) in the case of Dy and Ho and perpendicular to the c -axis ($\mathbf{P}_\mu \perp \mathbf{c}$) for Er.

In all the three metals, spontaneous muon spin precession was observed in both the ferromagnetic and the antiferromagnetic states. The temperature dependences of the spin rotation frequencies $\nu_\mu(T)$ are shown in Figs. 1–3 for Dy, Ho and Er, respectively.

Within the afm regimes we observe a smooth Brillouin-like increase of precession frequency with

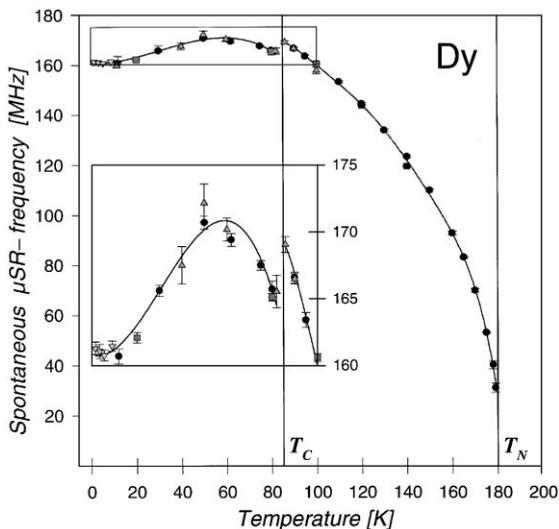


Fig. 1. Temperature dependence of the spontaneous muon precession frequency $\nu_\mu(T)$ in the ferromagnetic ($T < T_C \approx 86$ K) and helical antiferromagnetic ($T_C < T < T_N \approx 180$ K) temperature regime of single-crystalline dysprosium.

decreasing temperature. The fit to the data below T_N using a power law $B_\mu(T) \propto (T_N - T)^\beta$ allows the determination of the antiferromagnetic ordering temperatures. They are listed in Table 1. Small discontinuities (3% and 7% respectively) are visible at the ferromagnetic transition $T_C \approx 86$ K in Dy and at the basal ordering temperature $T_{N,\perp} \approx 53$ K in Er. There is no indication of a c -axis moment in Dy below $T < 10$ K as proposed by Willis et al. [2]. Irregularities in precession frequency at the proposed ‘spin-slip-transition’ temperatures in Ho and Er were not seen.

Most curious – and still unexplained – is the observation that the frequencies *decrease* in the ferromagnetic state as $T \rightarrow 0$. Since this effect is seen in all three investigated metals with a similar shape and order of magnitude ($\sim 10\%$) we must conclude that it is a general feature which is induced and/or exclusively seen by the muon as a local probe. The simplest explanation would be a slight change of cone angle with temperature. Dipolar field calculations, however, cannot reproduce the observed effect with reasonable input parameters. In particular, the straightforward ferromagnetic structure of Dy excludes such an

Table 1

Saturation values ($T \rightarrow 0$) of the measured local magnetic field B_μ , the calculated dipolar field B_{dip} and the extracted Fermi contact field $B_{\text{fc}} = B_\mu - B_{\text{dip}}$ at the octahedral interstitial site in single-crystalline samples of Gd, Dy, Ho and Er. The extrapolated values of B_μ and B_{fc} correspond to a Brillouin-like continuation of the afm temperature dependence of $B_\mu(T)$. The fit of the data to a power law $B_\mu \propto (T_N - T)^\beta$ determines the Néel temperatures T_N whereas the values of the Curie temperatures T_C were taken from Ref. [1]. Assuming instead the tetrahedral interstitial site as the muon position leads only to minor changes

	Saturation values ($T \rightarrow 0$)					T_C (K)	AFM transition	
	B_μ [T]		B_{dip} [T] Calcul.	B_{fc} [T]			$B_\mu \propto (T_N - T)^\beta$	
	Measured	Extrap.		Meas.	Extrap.		T_N (K)	β
Gd	0.110 (2)		0.889	-0.747		293	no AFM order	
Dy	1.186 (3)	1.34	1.321	-0.135	+0.02	85	180.5 (7)	0.41 (1)
Ho	1.563 (6)	1.73	1.244	+0.316	+0.49	20	131.2 (3)	0.46 (3)
Er	0.428 (1)	(2.86)	1.042	-0.622		20	86.6 (1)	0.46 (1)

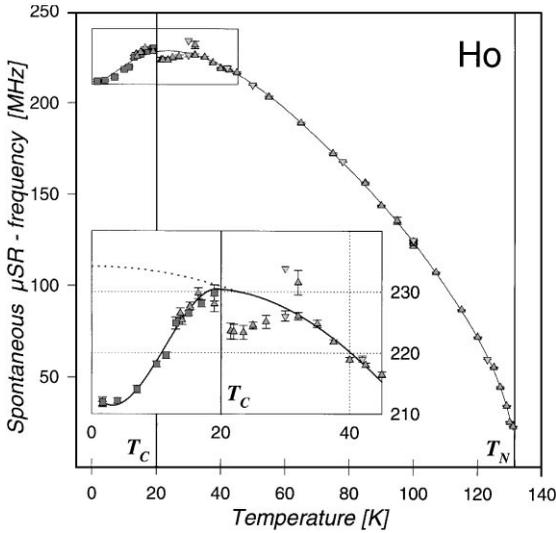


Fig. 2. Temperature dependence of the spontaneous muon precession frequency $\nu_\mu(T)$ in the conical ferromagnetic ($T < T_C \approx 20$ K) and helical afm ($T_C < T < T_N \approx 131$ K) temperature regime of single-crystalline holmium.

explanation if one assumes that the same mechanism is responsible in all three metals. This leaves a possible but still speculative explanation that – because of the strong magnetic anisotropy – bulk magnetization is not zero, even under zero external field cooling through the transition. The reduction of the local field is then the result of the presence of a demagnetizing field. The saturation values of the

local magnetic field $B_\mu(T \rightarrow 0)$ measured and extrapolated (from the Brillouin-type afm behavior) are compiled in Table 1.

The dramatic 80% drop of the spontaneous frequency $\Delta\nu_\mu(T_C) = (\gamma_\mu/2\pi) \cdot \Delta B_\mu(T_C)$ or of the local magnetic field $\Delta B_\mu(T_C) \approx 2.2$ T at the ferromagnetic transition $T_C \approx 20$ K of Er can be explained by a change of both the orientation and the magnitude of the dipolar field $B_{\text{dip}}(T_C)$ as illustrated in Fig. 3: The ferromagnetic orientation of the formerly antiparallel ‘domains’ each consisting of four parallel axial moments, produces a dipolar field $B_{\text{dip}}(T < T_C)$ of half the value and oppositely directed to that present in the antiferromagnetic regime. The relatively small contact field B_{fc} shows no change in either orientation or magnitude, and enhances the drop of the local field $\Delta B_\mu(T_C)$ at the muon site. The existence of two spontaneous rotation signals with comparable amplitudes in the ferromagnetic range of Er is a clear indication for two equally distributed, magnetically different muon environments and has been previously reported and discussed [3].

A coexistence of two precession frequencies is also visible in Ho, but now in the temperature regime $T_C < T < 35$ K, where the helix becomes more and more distorted by ‘spin-slips’ and – as a direct consequence – different magnetic muon environments appear. One of the two signals disappears before entering the ferromagnetic conical structure below $T_C \approx 20$ K. Dipolar calculations

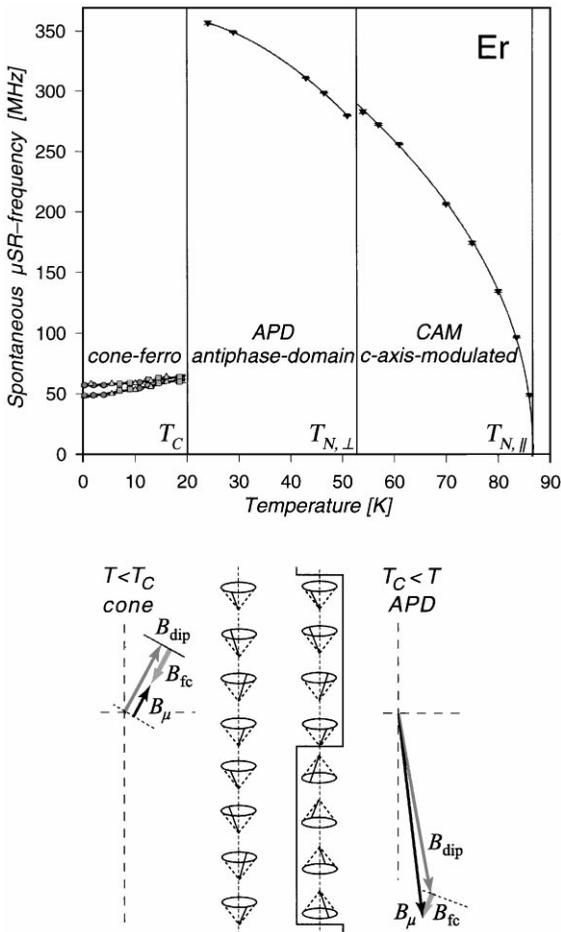


Fig. 3. Temperature dependence of the spontaneous muon precession frequency $\nu_\mu(T)$ in the conical ferromagnetic ($T < T_C \approx 20$ K) and both afm temperature regimes (APD: $T_C < T < T_{N,\perp} \approx 53$ K and CAM: $T_{N,\perp} < T < T_{N,\parallel} \approx 86$ K) of single-crystalline erbium.

clearly show that a regular bunching of the basal moments along the b -axis produces a single frequency consistent with our experimental results below T_C .

Due to the equal distribution of magnetic domains in zero external field the standard μ SR rotation measurements allow the determination of the frequency but not the sense of the muon precession. In general then, there are two (mathematical) solutions for the contact field B_{fc} depending on the

relative orientation of the measured local fields B_μ and the calculated dipolar field B_{dip} . Pronounced features in the measured local fields $B_\mu(T)$ in Gd (i.e. its unusual temperature dependence) and Er allows to select one of the solutions based on the changes of their axial moment components. The same approach is not possible for Dy and Ho where the magnetic moments are mainly confined to the basal plane and the dipolar anisotropy has no effect. Additional measurements had to be performed in an external magnetic field, which produces a rather small, but non-vanishing, sample magnetization. The comparison of the signals from longitudinal and transverse detector arrangements allowed the determination of the sense of spin rotation. The result is consistent with a parallel orientation of the local magnetic field relative to the sample magnetization ($B_\mu \parallel M$). This in turn leads to the values of the magnitude of the contact field B_{fc} listed in Table 1.

In contrast to Gd, the dipolar field at the octahedral and tetrahedral interstitial site are of similar size and shape in Dy, Ho and Er and so we are not able to decide between the two possible muon sites at present. It is hoped that the discontinuities of $B_\mu(T)$ observed at T_C in Dy or at $T_{N,\perp}$ in Er and the ‘splitting’ of the signals at $T \approx 30$ K in Ho, together with theoretical estimations of the conduction electron polarization, will help to solve this problem in the not too distant future.

Acknowledgements

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References

- [1] J. Jensen, A.R. Mackintosh, Rare Earth Magnetism – Structures and Excitations, Clarendon Press, Oxford, 1991.
- [2] F. Willis, N. Ali, J. Phys.: Condens. Mater. 2 (1990) 8205.
- [3] O. Hartmann, E. Lidström, M. Ekström, R. Wäppling, L. Asch, G.M. Kalvius, Hyperfine Interactions 104 (1997) 293.