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Evidence for perpendicular magnetic anisotropy of Tb in Tb/Fe multilayers

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Abstract. The magnetic moment direction of Tb in an [Fe(40 Å)/Tb(30 Å)]$_3$ multilayer has been studied by the low-temperature nuclear orientation technique in external magnetic fields applied along and perpendicular to the sample plane. A perpendicular magnetic anisotropy of the Tb magnetic moments has been found at zero external magnetic field and the mean misalignment angle has been estimated to be about 49° with respect to the sample plane normal. The external magnetic field applied along the sample plane turned the Tb magnetic moments to the plane, without reaching any saturation even at $B_{ext} = 8$ T. A possible structure of misalignment within the Tb layer is discussed.

1. Introduction

Fe/rare-earth (RE) multilayers show some unusual magnetic properties (see, e.g., [1–5]) and are potential candidates as magneto-optical recording media (see, e.g., [6]). Their most attractive feature is the perpendicular magnetic anisotropy (PMA) which has been found for Fe/RE with RE = Pr, Nd, Tb, Dy or Eu by magnetization and ^{57}Fe Mössbauer spectroscopy studies [1–5]. Although the RE atoms are believed to play a crucial role in the PMA mechanism [2, 5], direct information on the RE magnetic moment direction structure in Fe/RE with PMA, which is available for Fe [4, 5], is missing and the origin of PMA in Fe/RE multilayers is still open to discussion. To our knowledge, only neutron diffraction studies of Fe/Dy [7] and Fe/Nd [8] and a magnetic circular x-ray dichroism study of Fe/Tb [9] could give some support to the fact that RE magnetic moments are aligned perpendicular to the multilayer surface.

The low-temperature nuclear orientation (NO) technique (see, e.g., the review in [10]) can, in principle, give information on the local directions of chosen magnetic moments at low temperatures (i.e. their ground state in most cases). As this technique can unambiguously distinguish the moments of a chosen element from other moments in a given system, it can be very helpful in the study of magnetism of complicated composite materials such as

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multilayers. However, at least to our knowledge, the only NO study of magnetic multilayers yet published is the study of ^{110m}Ag in Ag/Fe multilayers (the first results were given in [11]).

Here we present the first results of our NO study of the Tb moment behaviour in an [Fe(40 Å)/Tb(30 Å)]$_{30}$ multilayer.

2. Experiment

The [Fe(40 Å)/Tb(30 Å)]$_{30}$ multilayer was prepared by alternate deposition in ultrahigh vacuum (10^{-9} Torr range) on a polymide film that was kept at $-50\,^\circ$C to prevent an intermixture at the interfaces. A Cu layer with a thickness of 3000 Å was then evaporated onto the multilayer to protect it and to enable soft soldering to the cold finger of a refrigerator. Stable ^{159}Tb was then activated to radioactive ^{160}Tb (half-life, 72 d) by irradiation of the sample in a reactor, in an Ar atmosphere and at temperatures below $80\,^\circ$C. The total neutron dose was about 2×10^{18} cm^{-2}.

The NO experiments were carried out using the Leuven and Prague NO facilities in the temperature range 7–50 mK. Measurements were performed with the external magnetic field B_{ext} both parallel to the sample plane (B_{ext} up to 8 T) and perpendicular to it (B_{ext} up to 3.5 T). The anisotropy W of the 299 keV ^{160}Tb γ line was monitored with a pure Ge detector placed along the B_{ext} direction (0° detector). When B_{ext} is parallel to the sample plane, another detector was placed in the direction perpendicular to the sample plane (90° detector, available only for $B_{ext} \leqslant 1$ T) A ^{60}Co:Co single crystal and ^{54}Mn:Ni NO thermometers were used.

The magnetization of the multilayer sample was also measured (before its activation) with a SQUID magnetometer at 5 K and at B_{ext} up to 5 T.

3. Experimental results and their analyses

An example of the temperature dependence of W ($B_{ext} = 1$ T, parallel geometry) is shown in figure 1. The B_{ext} dependence of W (temperature, around 10 mK) for the parallel geometry is given in figure 2 and for the perpendicular geometry in figure 3. The B_{ext} dependence of the magnetization at 5 K (B_{ext} both parallel and perpendicular to the sample plane) is shown in figure 4.

The anisotropy W of the 299 keV ^{160}Tb γ line is described by the formula (see, e.g., [10])

$$W = 1 + Q_2 U_2 A_2 B_2(B_{hf}, V_{zz}, T)\langle P_2(\cos\phi)\rangle \tag{1}$$

where Q_2, U_2 and A_2 (B_2) are the known NO constants (function), B_{hf} and V_{zz} are the hyperfine field and the electric field gradient, respectively, acting on the ^{160}Tb nucleus, P_2 is the Legendre polynomial (i.e. $(3\cos^2\phi - 1)/2$) and ϕ is the angle between the detector axis and the hyperfine interaction quantization axis. One can suppose that the Tb hyperfine interaction parameters in our system are similar to the Tb free-ion parameters, where the orbital momentum dominates, so that to a good approximation the direction of the quantization axis coincides with the Tb magnetic moment direction.

To demonstrate that the hyperfine interaction parameters of Tb in our multilayer are close to those of Tb in bulk Tb, we fitted the temperature dependence of W (see figure 1) with three free parameters, i.e. B_{hf}, V_{zz} and $\langle P_2\rangle$, using (1) and considering $U_2 A_2 = -0.365$ [12] and $Q_2 = 0.974$ (parallel geometry; 90° detector). The best fit resulted in $B_{hf} = 318(20)$ T, $V_{zz} = 33(6) \times 10^{21}$ V m^{-1} and $\langle P_2\rangle = 0.128(5)$ (the full curve in figure 1) to be

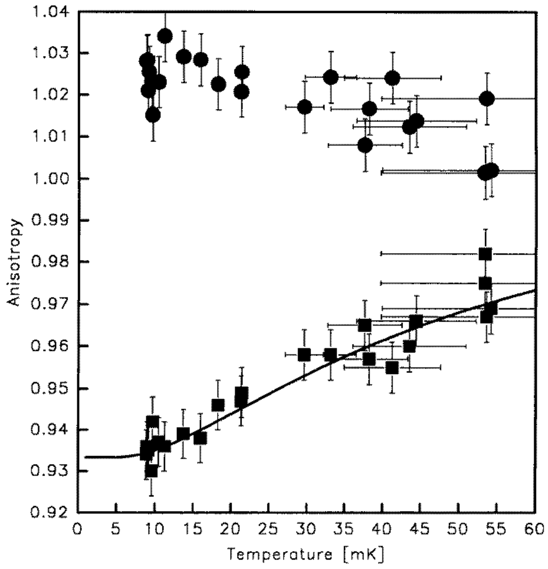

Figure 1. The temperature dependence of W for B_{ext} parallel to the sample plane and $B_{ext} = 1$ T: ●, 0° detector; ■, 90° detector; ——, best fit (see text).

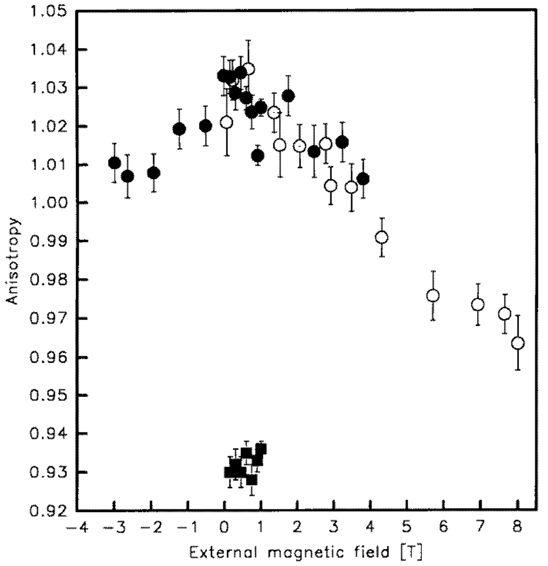

Figure 2. The external magnetic field dependence of W for B_{ext} parallel to the sample plane around 10 mK: ○, B_{ext} sweep down (0° detector); ●, B_{ext} sweep up (0° detector); ■, B_{ext} sweep up (90° detector).

compared with the hyperfine interaction parameters for bulk Tb, i.e. $B_{hf} = 299.3$ T and $V_{zz} = 40.8 \times 10^{21}$ V m^{-1} [13]. The Tb magnetic moment in our multilayer can therefore be supposed to hold its saturation free-ion value of $9\mu_B$.

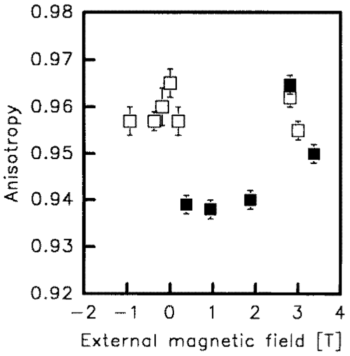

Figure 3. The external magnetic field dependence of W for B_{ext} perpendicular to the sample plane around 10 mK: ■, B_{ext} sweep up; □, B_{ext} sweep down.

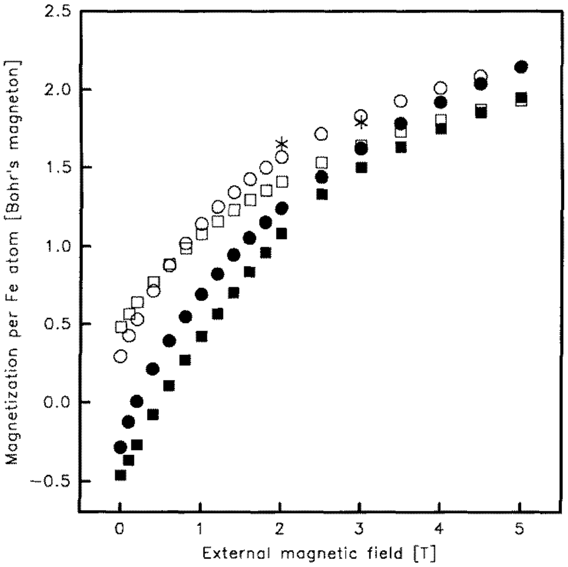

Figure 4. The external magnetic field dependence of the magnetization at 5 K: ●, B_{ext} sweep up (B_{ext} parallel to the sample plane); ○, B_{ext} sweep down (B_{ext} parallel to the sample plane); ■, B_{ext} sweep up (B_{ext} perpendicular to the sample plane); □, B_{ext} sweep down (B_{ext} perpendicular to the sample plane); *, values obtained from the combination of NO and Mössbauer spectroscopy (B_{ext} parallel to the sample plane normal) (see text).

Similarly to bulk Tb, the anisotropy W (in fact B_2) is almost saturated below 15 mK (see figure 1) and depends only on $Q_2U_2A_2$ and $\langle P_2\rangle$ in this temperature range. Nevertheless, to extract more accurate information on the Tb magnetic moment direction, which can be obtained from $\langle P_2\rangle$, we recalculated the low-temperature W-values from figures 2 and 3 for $\langle P_2\rangle$, using proper U_2A_2 and Q_2. The resulting B_{ext} dependences of $\langle P_2\rangle$ are shown in figures 5 and 6.

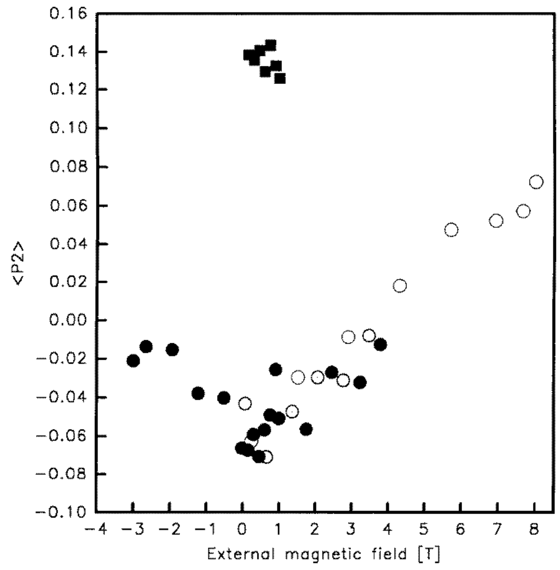

Figure 5. The external magnetic field dependence of $\langle P_2 \rangle$ for B_{ext} parallel to the sample plane: ○, B_{ext} sweep down (0° detector); ●, B_{ext} sweep up (0° detector); ■, B_{ext} sweep up (90° detector).

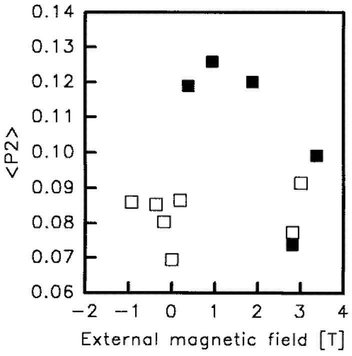

Figure 6. The external magnetic field dependence of $\langle P_2 \rangle$ for B_{ext} perpendicular to the sample plane: ■, B_{ext} sweep up; □, B_{ext} sweep down.

4. Discussion

In the limit of low B_{ext}, one can suppose symmetry around the plane normal. For the Tb magnetic moments fully oriented along this normal we should have $P_2(90^\circ \text{ detector}) = 1$ and $P_2(0^\circ \text{ detector}) = -\frac{1}{2}$. On the other hand, for the Tb magnetic moments randomly lying in the plane we should have $P_2(90^\circ \text{ detector}) = -\frac{1}{2}$ and $P_2(0^\circ \text{ detector}) = \frac{1}{4}$. Our values, i.e. $\langle P_2(90^\circ \text{ detector})\rangle = 0.14$ and $\langle P_2(0^\circ \text{ detector})\rangle = -0.07$ (see figure 5), follow the same symmetry (the ratio of $P_2(90^\circ \text{ detector})/P_2(0^\circ \text{ detector})$ should be -2) and, simultaneously, they are closer to the former case than to the latter case. This can be taken as evidence that the Tb magnetic moments are turned out of the sample plane and show PMA.

To interpret the $\langle P_2 \rangle$-values in detail, let us consider the following extreme cases.

(a) All Tb magnetic moments have the same misalignment with respect to the plane normal (we cannot straightforwardly give $\langle \phi \rangle$ because $\langle P_2(\cos\phi) \rangle \neq P_2(\cos\langle \phi \rangle)$ in general). In this case we obtain that the Tb magnetic moment direction is at angle of 49° with respect to the plane normal (for $B_{ext} \to 0$). This is somewhat higher than but comparable with the value of 42° found by Mössbauer spectroscopy for the Fe magnetic moments in the same Fe/Tb multilayer [5].

(b) Some Tb magnetic moments are fully perpendicular to the plane and the rest are randomly oriented. In this case we obtain for the fraction of the fully perpendicular Tb magnetic moments the value 14%. This could mean for example 2.1 Å on each side of the Tb layer in our multilayer, which is somewhat less than the estimated mean Tb monolayer thickness (about 3.7 Å).

When B_{ext} applied along the sample plane, is increased, the Tb magnetic moments smoothly turn into the sample plane, which is seen in figure 5 as an increase in $\langle P_2(0^\circ \text{ detector}) \rangle$ (and a decrease in $\langle P_2(90^\circ \text{ detector}) \rangle$ for $B_{ext} \leqslant 1$ T). The Tb mangetic moments are far from saturation even at $B_{ext} = 8$ T.

The data taken in the perpendicular geometry (B_{ext} along the plane normal) are in full coincidence with the parallel-geometry data at $B_{ext} \to 0$ (figure 3 and 6; B_{ext} sweep up). On the other hand, one can observe a certain decrease (!) in $\langle P_2 \rangle$ when B_{ext} is larger. This could be a demonstration of the fact that the Tb magnetic moments are originally oriented against B_{ext}, because they should be coupled antiparallel to the Fe layer, which has a larger magnetization than the Tb layer. (Because of the up–down symmetry this antiparallel orientation is not recognizable from a parallel orientation by γ-ray NO.) When B_{ext} increases, the Tb magnetic moments start to turn into the B_{ext} direction, which leads to a decrease in $\langle P_2 \rangle$ at the beginning of this process.

Simultaneously, one can find some signs of hysteresis in the B_{ext} dependence of $\langle P_2 \rangle$ in the perpendicular geometry (figure 6), which is not well understood and yet deserves further studies. This effect also has some support from the magnetization measurements, where the hysteresis with B_{ext} applied along the plane normal is considerably larger than that with B_{ext} applied along the plane (see figure 4).

We have tried to combine partial magnetizations, given by our NO results for the Tb layer and by the Mössbauer spectroscopy measurements for the Fe layer in the similar system Fe(40 Å)/Tb(26 Å) [4], and to compare such a result with the total magnetization (figure 4). Let us consider the case of B_{ext} along the plane normal and $B_{ext} = 2$ T. From [4] we have $\langle \phi \rangle = 23^\circ$ which leads to $\langle \mu_{Fe} \rangle = 1.97\mu_B$/Fe atom (we have used $\mu_{Fe,sat} = 2.14\mu_B$ obtained by Mibu [14] for a multilayer with the same composition). From our NO experiment we have $\langle P_2 \rangle = 0.12$ for Tb (figure 6). Using the extreme case (a) (see above) we obtain $\langle \mu_{Tb} \rangle = 1.55\mu_B$/Fe atom. One can see that neither parallel nor antiparallel combinations of $\langle \mu_{Fe} \rangle$ and $\langle \mu_{Tb} \rangle$ (i.e. $3.52\mu_B$/Fe atom and $0.42\mu_B$/Fe atom) agree with the total magnetization (figure 4). On the other hand, using the extreme case (b), and the presumption that the fully aligned Tb magnetic moments are already turned somewhat by B_{ext} (their local P_2 has changed from 1 to 0.86 to fulfil $\langle P_2 \rangle = 0.12$), we have $\langle \mu_{Tb} \rangle = 0.32\mu_B$/Fe atom. One can see from figure 4 that its antiparallel combination with $\langle \mu_{Fe} \rangle$ (i.e. $1.65\mu_B$/Fe atom) agrees with the total magnetization rather well.

The same comparison for B_{ext} along the plane normal and $B_{ext} = 3$ T (from [4], we have $\langle \phi \rangle = 18^\circ$ for Fe and, from NO, $\langle P_2 \rangle = 0.075$ for Tb) leads to $0.54\mu_B$/Fe atom for case (a) and $1.79\mu_B$/Fe atom for case (b), (only the antiparallel combination of $\langle \mu_{Fe} \rangle$ and

$\langle\mu_{Tb}\rangle$ is considered). Again, as one can see from figure 4, the latter case is rather close to the total magnetization value. Even the increase from $B_{ext} = 2$ to 3 T is kept.

The above comparison is of course very rough and many parameters are not yet clear enough. For example, the multilayer Tb/Fe measured by Mössbauer spectroscopy in [4] is not completely identical with ours. It is also not clear whether only the Tb magnetic moments, originally fully aligned, are turned by B_{ext}, as was considered for case (b), or whether there is some turning of originally randomly oriented Tb magnetic moments. Nevertheless, this comparison gives satisfactory agreement between the experimental results obtained by different techniques and supports the conclusion that case (b) (i.e. that only about 14% of the Tb magnetic moments is polarized) is closer to reality than case (a) (i.e. the same misalignment for all Tb magnetic moments). To verify and/or complete this picture more experiments with other Tb thicknesses are necessary and they are in progress now.

5. Conclusions

The NO orientation study of ^{160}Tb in the Fe(40 Å)/Tb(30 Å) multilayer showed that the Tb magnetic moments have perpendicular magnetic anisotropy and a mean angle of 49° with respect to the sample plane normal at $B_{ext} \rightarrow 0$. B_{ext} applied along the sample plane turns the Tb magnetic moments along its direction, but they are far from their saturation even at $B_{ext} = 8$ T. The comparison of the NO, magnetization and Mössbauer spectroscopy experiments supports the suggestions that PMA of the Tb magnetic moments is not unique through the whole Tb layer but is more pronounced for the Tb atoms lying close to the interface with the Fe layer.

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