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1998 J. Phys.: Condens. Matter 10 7467

(http://iopscience.iop.org/0953-8984/10/33/015)

J. Phys.: Condens. Matter **10** (1998) 7467–7474. Printed in the UK PII: S0953-8984(98)90757-3

Tb magnetic moment behaviour in a Gd single crystal: nuclear orientation study

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Received 15 January 1998, in final form 20 May 1998

Abstract. Low-temperature nuclear orientation has been used to study the behaviour of the Tb magnetic moment in a Gd + 1 at.% Tb single crystal. The γ-ray anisotropy of several ^{160}Tb lines was monitored in the temperature range of 5–50 mK and in the external magnetic field (B_{ext}) range of 0–8.5 T. The field was applied either along the a- or along the c-axis of the Gd single crystal. In the case of B_{ext} parallel to the a-axis the Tb magnetic moment was found to be almost aligned along B_{ext} even at very low fields, as expected because the hexagonal basal plane is an easy magnetization plane for the Tb ion in terbium metal. On the other hand, when B_{ext} is along the c-axis the Tb magnetic moment rotates very slowly towards the c-axis. Even at $B_{ext} = 8.5$ T the Tb magnetic moment is not along the direction of B_{ext}. This is in contrast to the fast saturation of the gadolinium ions in pure Gd. Apparently, the Tb–Gd exchange interaction is in competition with the crystal field acting on the Tb ion in a hexagonal lattice, our data indicates that at $B_{ext} = 8.5$ T along the c-axis the angle between the Tb magnetic moment and the applied field is of the order of 30°.

1. Introduction

Some rare earth (RE) metals show very peculiar magnetic structures, given in general by the interplay of the oscillatory exchange interaction of RKKY type and the crystal field. The magnetic structure of RE alloys may be even more complicated. To understand such magnetic structures, information about the behaviour of individual magnetic sublattices is important. Unfortunately, such information is rather scarce, because it is not easy to obtain experimentally.

The Tb**Gd** single crystal seems to be one of the simplest systems for such investigation as one of the constituents (i.e. Gd) exhibits only a negligible magnetocrystalline anisotropy, while the other (Tb) should be forced by the hexagonal matrix crystal field to have its magnetic moment perpendicular with respect to the crystallographic c-axis. The experimental study of this system is not yet very advanced and, moreover, the results of different methods partly disagree with each other. To our knowledge only the following studies of the Tb**Gd** system exist: (i) the torque magnetometer measurements

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0953-8984/98/337467+08$19.50

of Gd + 1.7 at.% Tb [1], where the behaviour of the Tb magnetic moment sublattice was derived by comparison with the measurement of pure Gd, (ii) the ^{155}Gd Mössbauer study [2], which revealed the properties of the Gd sublattice influenced by Tb, and (iii) the ^{159}Tb nuclear magnetic resonance (NMR) study [3] giving the behaviour of the Tb sublattice based on assumptions of hyperfine interaction properties.

One of the methods, which can give direct information of the magnetic behaviour of individual magnetic sublattices, is low-temperature nuclear orientation (NO). In principle, it can yield (via hyperfine interactions) both the direction and the magnitude of the *ionic* magnetic moments of a given sublattice in their ground state ($T \rightarrow 0$) (see, e.g., review [4]). In our previous work we used NO with ^{160}Tb as a probe to study the Tb**Gd** single crystal system [5]. The main aim of that study was to find the direction of Tb ionic magnetic moments in a Gd host at different magnitudes and orientations of external magnetic field (B_{ext}). That study was partly motivated by the necessity to clear up solid state aspects of those NO experiments where polycrystalline Gd was used as a host for the orientation of short-lived rare earth isotopes to obtain their nuclear decay and hyperfine interaction parameters and where a presumption of full Tb magnetic moment alignment along B_{ext} was used for the derivation of some parameters in certain cases (see, e.g., review [6]). The experimental results obtained in [5] sharply disagreed with the simple theoretical model and in our present opinion the discrepancy found may have been caused by degradation of the Gd single crystal by ^{160}Tb activity diffusion and/or by incorrect orientation of the Gd single crystal.

In this paper we present our new NO study of ^{160}Tb in the Gd + 1 at.% Tb single crystal prepared by a different method than we used in [5]. We have used the same samples, which had already been studied by NMR (partly published in [3]). Moreover, a combination of the NO and NMR results allows us to derive directly the Tb magnetic moment direction for different magnitudes and orientations of B_{ext}.

2. Experimental

Gd + 1 at.% Tb single crystals were prepared by Dr S Abell from the School of Metallurgy at the University of Birmingham. Gd and Tb of 4N purity were inductively melted. A single crystal was grown by zone refinement under pure Ar atmosphere and annealed for 100 h at 1200 °C. The orientation of the single crystal was determined by Laue x-ray back-scattering. Two single crystalline Gd + 1 at.% Tb samples with the shape of a needle (length ∼1 cm, diameter ∼1 mm) along the *a*- or *c*-axis were then cut by spark erosion. Their final orientation was again checked by Laue x-ray back-scattering and was found to be better than one degree. In order to remove any damage to the surface during the spark erosion process, the needles were slightly etched in dilute nitric acid. These samples were identical to ones studied in [3] by high frequency NMR.

To obtain radioactive ^{160}Tb nuclei in the samples (half-life 72 days), these were irradiated in a reactor with thermal neutrons.

The NO experiments were carried out using the Leuven and Prague NO facilities in the temperature range 5–50 mK and in the B_{ext} range 0–8.5 T, applied along the *a*- and *c*-axis of the corresponding samples. The γ-ray anisotropy W of several ^{160}Tb lines was monitored by a Ge detector placed along B_{ext} and analysed by standard NO methods (see, e.g., [4]). The reference measurements ('warm states') were taken at temperatures of 1.4–4.2 K, where the γ-ray radiation of ^{160}Tb should be isotropic ($W = 1$). A ^{54}MnNi NO thermometer was used for the temperature determination.

3. Results and analysis

The temperature dependence of W was measured for both orientations of B_{ext} (i.e. along the a- or c-axis of the Tb**Gd** single crystal) and for several values of B_{ext}. In addition, the magnetic field dependence of W was measured for both orientations of B_{ext} as well, keeping the temperature below 9 mK.

Figure 1 shows some examples of the temperature dependence of W for the 299 keV ^{160}Tb γ-line. This line is the most intense in the decay of ^{160}Tb and its anisotropy W should show a large effect, the description of which is simpler than for other ^{160}Tb γ-lines, because the anisotropy coefficient $A_4 = 0$ for this line [7]. This 299 keV γ-line is therefore the most suitable one for further analyses. Its anisotropy W is described by the formula [4]

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

where Q_2, U_2, A_2 (B_2) are the known NO constants (function), B_{hf} and V_{zz} are the hyperfine field and the electric field gradient acting on the ^{160}Tb nucleus, respectively, P_2 is the Legendre polynomial (i.e. $(3\cos^2\phi - 1)/2$), and ϕ is the angle between the detector axis (= the B_{ext} direction) and the hyperfine interaction quantization axis. To an adequate approximation the latter axis is collinear with the Tb ionic magnetic moment, because in the case of Tb the ionic part of the electric field gradient dominates over the lattice one and its symmetry axis coincides with the hyperfine field and ionic magnetic moment directions (see, e.g., [8]).

In order to obtain from equation (1) the direction of the Tb magnetic moment (i.e. $P_2(\phi)$) we use $Q_2 = 0.982$, given by our experimental geometry, and $U_2 A_2 = -0.365$ for the 299 keV ^{160}Tb γ-line [7]. Moreover, we use in the analysis the hyperfine interaction param-

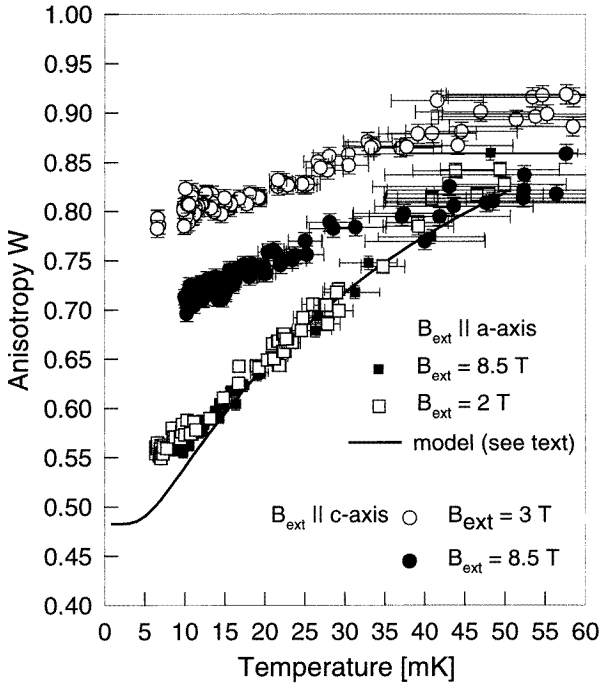

Figure 1. Temperature dependence of W of the 299 keV ^{160}Tb γ-line for different B_{ext} values and orientations.

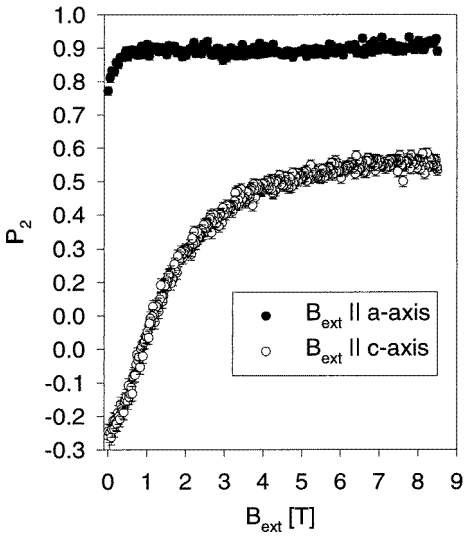

Figure 2. B_{ext} dependence of P_2 from the 299 keV ^{160}Tb γ-line.

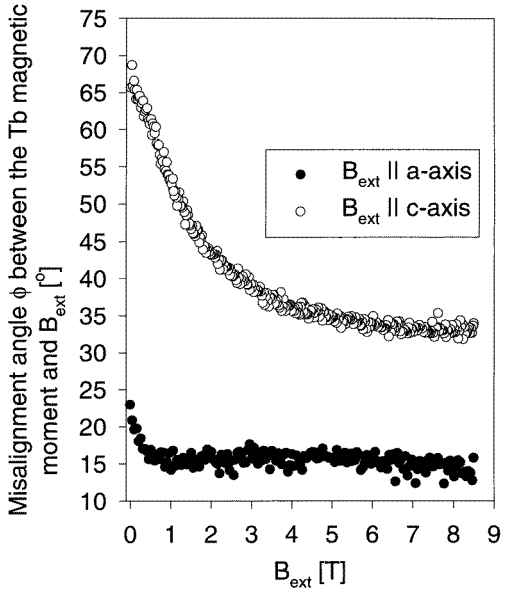

Figure 3. B_{ext} dependence of the misalignment angle ϕ of the Tb magnetic moment from the 299 keV ^{160}Tb γ-line.

eters determined in the NMR experiments on the same samples, corresponding to B_{ext} values and directions (partly published in [3]), and take into account the known magnetic dipole and electric quadrupole moments of the ^{159}Tb [9] and ^{160}Tb [10] nuclei. Thus, using equation (1) we could recalculate our experimental values of W for $P_2(\phi)$ and thus obtain information on the Tb magnetic moment direction. Figure 2 shows the B_{ext} dependence of $P_2(\phi)$ calculated from the experimental W-values measured at temperatures below 9 mK for both B_{ext} orientations. In figure 3 the corresponding ϕ-values, calculated from $P_2(\phi)$, are shown.

4. Discussion

Gadolinium metal is known to be magnetically very soft, but the Tb ion in a hexagonal crystallographic environment in the Gd matrix should have an easy magnetization plane parallel to the hexagonal basal plane (see, e.g., pure Tb single crystal [11]). Furthermore, from analogy with pure Tb or Gd systems one can suppose that among both Tb–Gd and Gd–Gd exchange interactions a ferromagnetic coupling prevails. From these facts we can derive that B_{ext} applied in the basal plane should very easily align the Tb magnetic moments along its direction (and along the total magnetization direction of the sample). Strong support for such a conclusion is also given by the NMR results [3], which simultaneously provide evidence that the Tb magnetic moments are parallel to B_{ext} (and not antiparallel as they could eventually be in the case of more complicated Tb–Gd exchange interactions). On the contrary, our older data [5] did not show such a feature which led us to doubt the quality of the samples that were used then.

Using equation (1) we can calculate W for the case where the Tb magnetic moments are parallel to B_{ext} (i.e. $P_2 = 1$) and the results for $B_{ext} = 2$ T are shown as a full curve in figure 1. One can see that this model agrees with our experimental points obtained for B_{ext} lying in the easy magnetization plane ($B_{ext} \parallel a$-axis) at higher temperatures, while there is a (small) disagreement between this model and our experimental data at temperatures below about 20 mK. A similar disagreement has also been found in the case of the other ^{160}Tb lines, i.e. 879 keV, 1178 keV, 1278 keV and also for the pure E2 966 keV transition, for which the values of the U_2A_2 and U_4A_4 coefficients are exactly known [4]. To clarify this deviation we have tried to fit our experimental temperature dependence of W, using equation (1), with B_{hf}, V_{zz} and P_2 as free parameters and supposing various models, which could explain the observed deviation. The first model assumed a misalignment of the Tb magnetic moments and reasonable values of hyperfine interaction parameters (i.e. deviating not more than $\pm 50\%$ from those of the Tb free ion), including a possible misalignment between the symmetry axes of the magnetic and electric hyperfine interaction. The second model assumed the existence of two different Tb atom positions, each with their own misalignment: one with the hyperfine interactions found by NMR and another with free hyperfine interaction parameters, but deviating no more than $\pm 50\%$ from the NMR ones. Unfortunately, no such model could fit our experimental data (measured several times with the same result at both Leuven and Prague facilities). It is interesting that a similar deviation was found in the study of ^{160}Tb nuclei in the pure Tb single crystal at temperatures around 6 mK [12], while a full agreement between theory and experiment was reported for temperatures above 18 mK [13]. This observation is identical to ours and is thought to have some deeper reason.

A possible explanation could be as follows: after activation of the Tb nucleus by a neutron ($^{159}\mathrm{Tb} + \mathrm{n} \rightarrow {}^{160}\mathrm{Tb}$) the ^{160}Tb nucleus is left in an excited state ($\sim$7 MeV), which is promptly de-excited to the ^{160}Tb ground state by emission of a γ-ray cascade (see the simulations of this process, e.g., in [14]). The individual γ-ray emissions of energies up to several MeV give knock-ons of energies up to several tens of eV to the Tb ion which can cause its displacement and/or create a vacancy, a Frankel pair or other lattice damage (see, e.g., [15]). This damage is rather rare in bulk (and therefore may bring only negligible change in bulk properties), but it is located near the position of the nucleus measured afterwards by NO (i.e. ^{160}Tb) and can therefore dramatically change the symmetry in the nucleus neighbourhood and therefore the hyperfine interactions acting on it. Unfortunately, no reliable simulations of this process (properly including both the nuclear de-exciting γ-ray cascade (like in [14]) and the subsequent motion of a given atom

(molecular dynamics computing)) are available yet. The question of whether this effect could cause the discrepancy found by us, remains unsolved.

On the other hand, our results (see figure 2) roughly coincide with our expectation that P_2 is saturated by rather low B_{ext} (<1 T) in the B_{ext}-in-basal-plane case. One can find that the saturation misalignment is not too large (∼15°—see figure 3) and in fact not too far from a full alignment. Nevertheless, we must emphasize that our data cannot be, as mentioned previously, explained only by a simple Tb magnetic moment misalignment caused, for example, by an experimental mismatch between B_{ext} and the crystallographic a-axis (proposed in [12]). One can only estimate roughly that at least 90% of the Tb ions may show the expected behaviour (i.e. the same hyperfine interaction parameters as the NMR measurements and the full alignment of the Tb magnetic moment), while less than 10% behave obscurely.

In the $B_{ext} \parallel c$-axis case we find a completely different behaviour. Indeed, B_{ext} only turns the Tb magnetic moments slowly from being almost perpendicular to the c-axis if $B_{ext} = 0$, to the B_{ext} direction (and thus to the c-axis) at higher B_{ext}. Nevertheless, the Tb magnetic moments are far from their full alignment along the c-axis even at $B_{ext} = 8.5$ T (see figures 2 and 3). This tendency (and even the values of P_2 and ϕ) is not much changed, even if one takes into account that only about 90% of the Tb ions behave properly (and the rest are completely misaligned) as concluded earlier. In this case the $P_2(\phi)$ values at $B_{ext} = 0$ and 8.5 T would only change from −0.25 (67°) and 0.55 (33°), to −0.29 (69°) and 0.61 (30°), respectively.

This behaviour, i.e. the very difficult saturation of the Tb magnetic moment in Gd in the c-axis direction, is at least in qualitative agreement with the conclusions based on the torque magnetometer measurements of Gd + 1.7 at.% Tb, where the Tb magnetic moment misalignment from the c-axis at $B_{ext} = 3.1$ T ($B_{ext} \parallel c$-axis) was found to be 16° [1] (our value of the Tb magnetic moment misalignment for the same B_{ext} value and orientation, but for the Gd+1 at.% Tb system, it was 38°). On the other hand, our results sharply contradict the conclusions based on the ^{159}Tb**Gd** NMR results where a lack of the B_{ext} dependence of the hyperfine parameters for $B_{ext} > 3$ T was taken as evidence for a full Tb magnetic moment alignment along the c-axis [3]. This discrepancy has initialized refined analyses of the older NMR data [3] considering the orientation of the Tb magnetic moment as a free parameter to be fitted to the NMR experimental data. These new analyses of the NMR measurements are still progressing and their full description will be published elsewhere [16]. However, the preliminary results of these analyses give a value of the misalignment between the Tb magnetic moment and the applied field along the c-axis in good agreement with that of the NO experiment (e.g. for $B_{ext} = 6$ T the Tb magnetic moment misalignment with respect to the c-axis is found to be about 30° from the NMR data, compared to about 33° from the NO measurements—see figure 3). Therefore, the simple assumption of full collinearity between the Tb and Gd magnetic moments made in [3] is not confirmed even from the NMR data themselves.

Let us now concentrate on the zero B_{ext} results. The results of both B_{ext} directions show that the Tb magnetic moment *does not lie* in the Gd crystallographic a-axis, as it was assumed in [3]. One can find that the sum of the Tb magnetic moment misalignment angles from the a- and c-axes (i.e. 23(2)° and 67(2)°, respectively) is just 90°. One can conclude that at zero B_{ext} the Tb magnetic moment does not lie in the crystallographic basal plane, as is the case for pure Tb [11], and that it has its direction approximately in the plane given by the a- and c-axes. This seems to be natural when one takes into consideration a mutual competition of (i) the Gd magnetic anisotropy, which fixes the Gd magnetic moments to be at an angle of 28° with the c-axis (e.g. [2]) and which is partly

transferred on the Tb magnetic moments via Tb–Gd exchange interaction, and (ii) the Tb ion anisotropy in a hexagonal lattice, which should force the Tb magnetic moment into the basal plane. At least qualitatively a similar feature was reported in [2], where the ^{155}Gd Mössbauer study yielded a Gd magnetic moment misalignment with respect to the *c*-axis of about 78° in the Tb**Gd** powder of the same composition as our sample. Taking into account the fact that the Tb magnetic moment misalignment should have been even larger, i.e. between this value and 90°, one can see that the agreement of our results and those in [2] is not quantitative. However, a sharp inconsistency between the single crystal and powder samples was reported in [2], therefore the misalignment value given in [2] may not be quite as reliable.

5. Conclusions

Using low-temperature nuclear orientation we have directly observed that the Tb magnetic moments in the Gd host do not follow the pure Gd magnetization direction even at high external magnetic fields (up to 8.5 T), when the field is applied along the Gd crystallographic *c*-axis. The dependence of the Tb magnetic moment misalignment on the external magnetic field has been found. This behaviour is probably caused by competition between the Tb–Gd exchange interaction, which forces the Tb magnetic moment into the *c*-axis direction, and the crystalline field of the hexagonal Gd host, which holds Tb magnetic moments in the basal plane. A full model, which would quantitatively describe the whole process should take into account the crystal field parameters and the Tb–Gd and Gd–Gd exchange parameters. This model is awaited but in progress.

Acknowledgments

One of the authors (MT) would like to thank the Academic Board of Leuven University for the award of a Research Fellowship, during which most of the experimental work was carried out. The authors also with to thank Dr S Abell from the University of Birmingham for the single crystals preparation. A part of this work was supported by the Grant Agency of the Czech Republic under contract No 202/94/0501.

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