



Spin Reorientation Transitions and Crystal Field Interactions in the $R(Fe_{10}V_2)$ Series

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Spin reorientation transitions to a ground state where the magnetization is not parallel to *c*-axis, are observed below room temperature in $R(Fe_{10}V_2)$, for $R = Nd, Dy, Ho$ and Er . These are quantitatively explained by using a set of five crystal-field coefficients derived from the fit of the magnetization curves of a $Dy(Fe_{11}Ti)$ crystal. An explanation is given of why first-order spin reorientation transitions are found in $Ho(Fe_{10}V_2)$ and $Ho(Fe_{10}Mo_2)$, but not in $Ho(Fe_{11}Ti)$.

INTRODUCTION

Interest in the magnetic properties of iron-rich rare-earth compounds with the $ThMn_{12}$ structure has been stimulated by the prospect of using the Sm alloys as permanent magnets. Many such compounds with general formula $R(Fe_{12-x}M_x)$ [1] have been synthesized with $M = Ti, V, Cr, Mo, W$ and Si where $1 \leq x \leq 3$. A study of spin reorientation transitions (S.R.T.) in the entire $R(Fe_{11}Ti)$ series [2], and a subsequent study of the magnetization curves of a $Dy(Fe_{11}Ti)$ crystal [3] have led to a complete set of five crystal-field coefficients: $A_{20} = -32.3 \text{ Ka}_0^{-2}$, $A_{40} = -12.4 \text{ Ka}_0^{-4}$, $A_{44} = 118 \text{ Ka}_0^{-4}$, $A_{60} = 2.56 \text{ Ka}_0^{-6}$, $A_{64} = 0.64 \text{ Ka}_0^{-6}$, and exchange field coefficients n_{RFe} which account for the intrinsic magnetic properties of the series. Notable is the small negative value of A_{20} , which means that, (i) The uniaxial iron sublattice anisotropy $K_1(Fe)$ is usually the dominant contribution at room temperature and above, leading to alignment of the magnetization along the *c*-axis regardless of the sign of the rare earth second-order Stevens coefficient α_J and (ii) anisotropy resulting from higher-order rare-earth crystal-field terms influences the magnetic

structures up to comparatively high temperatures ($\sim 150 \text{ K}$).

There are several reports of S.R.T. in the $R(Fe_{10}V_2)$ series [4-9]. $T_{SR} = 120 \text{ K}$ for $R = Nd$ [4,5], 200 K for $R = Dy$ [6] and 60 K for $R = Er$ [7-9]. For Nd and Er in the ground state the magnetization is directed at an angle of $\sim 45^\circ$ to the *c*-axis. Moze *et al* discussed the behaviour of the Er compound in a model with two rare-earth crystal-field coefficients A_{20} and A_{40} .

Here we study the S.R.T. for the $R(Fe_{10}V_2)$ series in more detail and use the crystal-field and exchange model developed for the $R(Fe_{11}Ti)$ to account for the results. Since the ground state magnetic structure of compounds with $R = Ho$ is finely balanced between a *c*-axis and an inclined ground state, we also include some data on the $Ho(Fe_{10}Mo_2)$ compound, and explain why $Ho(Fe_{10}V_2)$ and $Ho(Fe_{10}Mo_2)$ compounds exhibit a *first-order* S.R.T., whereas the $Ho(Fe_{11}Ti)$ compound does not.

RESULTS AND DISCUSSION

The $R(Fe_{10}V_2)$ alloys for $R = Tb, Dy, Ho$ and Er , and $Ho(Fe_{10}Mo_2)$, were made by arc

melting of the pure elements followed by an anneal at 850 °C in vacuum for one week. Thermomagnetic scans on oriented samples of the $R(\text{Fe}_{10}\text{V}_2)$ series and on a polycrystalline $\text{Ho}(\text{Fe}_{10}\text{Mo}_2)$ sample in an fixed applied field of 0.05 T are shown in Fig. 1 and Fig. 2, respectively. To estimate the tilting angle α , between the magnetization direction and the tetragonal c -axis. A simple model is used, which assumes a distribution of the c -axis around the oriented direction. Using the distribution function: $P(\theta) = [(n+1)/2\pi] \cos^n \theta$, as used earlier to analyse S.R.T. from Mössbauer spectra [10], we find

$$\frac{\langle M_{//} \rangle}{M_s} = \frac{(n+1)}{(n+2)} [\cos \alpha + C(n) \sin \alpha] \quad (1)$$

$$\frac{\langle M_{\perp} \rangle}{M_s} = \frac{(n+1)}{(n+2)} C(n) \cos \alpha \quad (2)$$

$$\frac{\langle M_{//} \rangle}{\langle M_{\perp} \rangle} = C(n)^{-1} + \tan \alpha \quad (3)$$

where the constant $C(n)^{-1} = (\pi/2)n!/(n-1)!!$ (n is odd number), and is equal to the ratio of $\langle M_{//} \rangle / \langle M_{\perp} \rangle$ when α vanishes. Using Eq. (3), the variations of the tilting angle α as a function of temperature were deduced from thermomagnetic data. The spin reorientation transition temperatures could be precisely determined, although the values of α may not be entirely reliable. Values obtained are

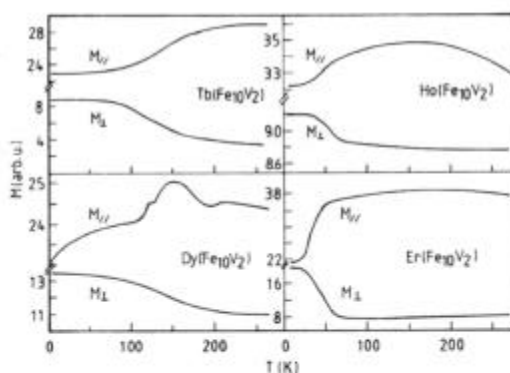


Fig. 1. Thermomagnetic data on $R(\text{Fe}_{10}\text{V}_2)$ ($R=\text{Tb}, \text{Dy}, \text{Ho}, \text{Er}$) compounds, obtained in an applied field of 0.05 T on oriented samples. $\langle M_{//} \rangle$ and $\langle M_{\perp} \rangle$ indicate the applied field parallel and perpendicular to the oriented direction respectively.

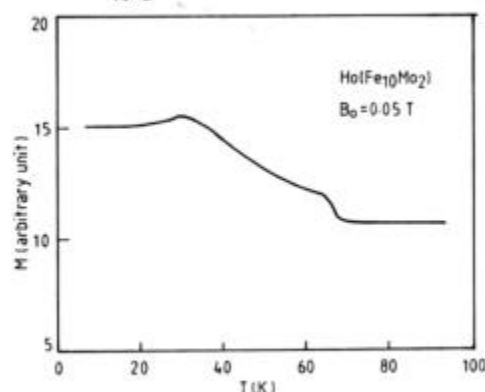


Fig. 2. Thermomagnetic curve on a $\text{Ho}(\text{Fe}_{10}\text{Mo}_2)$ polycrystalline ingot sample in an applied field of 0.05 T.

listed in the table 1. The S.R.T. occurring in the $R(\text{Fe}_{10}\text{V}_2)$ series are quite similar to those observed previously in the $R(\text{Fe}_{11}\text{Ti})$ series [2] except for $\text{Ho}(\text{Fe}_{11}\text{Ti})$, which shows no transition. However, in recent analysis of $\text{Dy}(\text{Fe}_{11}\text{Ti})$ single-crystal magnetization curves [3], the authors pointed out that a slightly weaker iron sublattice anisotropy could lead a first-order S.R.T. for other $\text{Ho}(\text{Fe}_{12-x}\text{M}_x)$ compounds.

Table 1. Curie temperatures, exchange coefficients and spin reorientation transition temperatures of $R(\text{Fe}_{10}\text{V}_2)$.

$R(\text{Fe}_{10}\text{V}_2)$	T_C (K)	μ_{RFe}^* (μ_B)	T_{SR}^{exp} (K)	T_{SR}^{cal} (K)
Nd	570	427	120	163
Tb	570	184	170	>300
Dy	540	175	140, 215	113, 262
Ho	525	186	60	76
Er	505	170	65	50

* The iron effective magnetic moment in the paramagnetic state was taken as $3.7 \mu_B$ [2].

The crystal-field and exchange interactions model described in references 2, 3 and 13, was used to explain the complex magnetic behaviour in the series. The full crystal-field Hamiltonian for the 2a site (point symmetry $4/mmm$) is

$$\begin{aligned} \mathcal{H}_{cf} = & B_{20}O_{20} + B_{40}O_{40} + B_{44}O_{44} \\ & + B_{60}O_{60} + B_{64}O_{64} \end{aligned} \quad (4)$$

The five crystal-field coefficients A_{nm} were taken to be those deduced for $\text{Dy}(\text{Fe}_{11}\text{Ti})$ [3]. The magnetization of the iron sublattice was determined from measurements for $\text{Y}(\text{Fe}_{10}\text{V}_2)$. The temperature dependence of the iron sublattice anisotropy $K_1(\text{Fe})$ for $\text{Y}(\text{Fe}_{10}\text{V}_2)$ was taken from the data of Solzi *et al* [11]. The exchange coefficients n_{RFe} were deduced from the values of T_c (see table 1), using the method described by Belorizky *et al* [12]. Then the S.R.T. temperatures, the magnetic structures and the energy surfaces were calculated. A value of n_{NdFe} of $325 \mu_0$ was taken in the calculation. The calculated temperature variations of the tilting angle α between the net magnetization and the c-axis is shown in Fig. 3. The agreement between experiment and calculation for the S.R.T. temperatures (see table 1) is quite good (except for Tb), in view of the fact that the crystal-field coefficients, found for $\text{Dy}(\text{Fe}_{11}\text{Ti})$ single crystal, have not been adjusted. Fig. 4 illustrates the energy surfaces in a (010) plane at 4.2 K for $\text{Ho}(\text{Fe}_{10}\text{V}_2)$ and $\text{Ho}(\text{Fe}_{11}\text{Ti})$, using exactly the same $\{A_{nm}\}$ in each case. It can be seen that two minima, one at $\alpha = 0$ and one at $\alpha \sim 55^\circ$, have a tiny difference in energy. The minimum at $\alpha = 0$ is lower for $\text{Ho}(\text{Fe}_{11}\text{Ti})$ and the one at $\alpha \sim 55^\circ$ is lower for $\text{Ho}(\text{Fe}_{10}\text{V}_2)$ or $\text{Ho}(\text{Fe}_{10}\text{Mo}_2)$. Hence no S.R.T. can be seen in $\text{Ho}(\text{Fe}_{11}\text{Ti})$, but there is a first-order transition in $\text{Ho}(\text{Fe}_{10}\text{V}_2)$ or $\text{Ho}(\text{Fe}_{10}\text{Mo}_2)$, resulting from the large energy barrier between the two minima. In the Tb case, the large discrepancy between the experimental and calculated results might be attributed to the admixture of different charge states of the Tb ion [3].

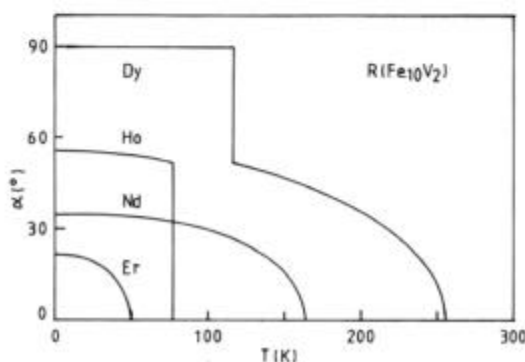


Fig. 3. Calculated temperature variation of the tilting angles for $R(\text{Fe}_{10}\text{V}_2)$ ($R = \text{Nd}, \text{Dy}, \text{Ho}, \text{Er}$).

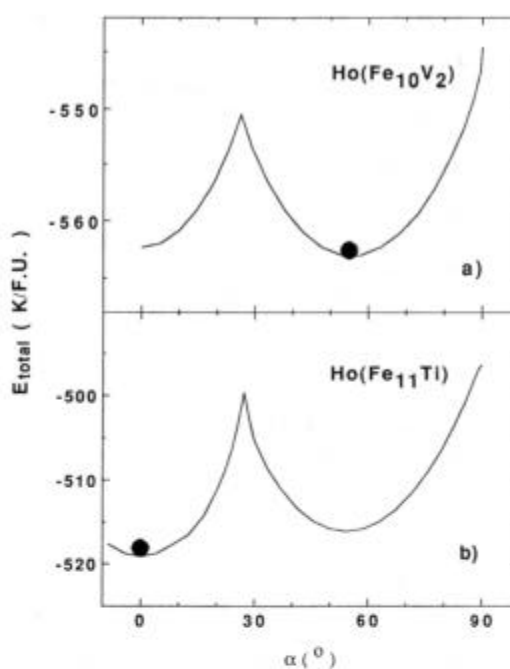


Fig. 4. Energy surfaces in the (010) plane at 4.2 K for a) $\text{Ho}(\text{Fe}_{10}\text{V}_2)$ and b) $\text{Ho}(\text{Fe}_{11}\text{Ti})$. α is the tilting angle between the net magnetization direction and the c-axis.

CONCLUSIONS

The complex S.R.T. appearing in the $R(\text{Fe}_{10}\text{V}_2)$ series can be well explained using a set of crystal-field coefficients deduced from measurements on the $\text{Dy}(\text{Fe}_{11}\text{Ti})$ crystal. In particular, the presence or absence of a first-order S.R.T. for $R = \text{Ho}$ emerges naturally from our model. We have confirmed that it is necessary to include all the crystal-field terms in the analysis to obtain the correct ground state magnetic configuration for the whole rare-earth series.

We conclude also that there is little change in the crystal-field coefficients for different transition metals M , at least when $x \leq 2$. However, there may be appreciable changes when the composition of the ThMn_{12} structure alloy is substantially different, as in $\text{Er}(\text{Mn}_4\text{Al}_8)$ [14] or $\text{Gd}(\text{Fe}_6\text{Al}_6)$ [15].

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