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Magnetic properties of rare-earth compounds of the $R\text{Co}_{10}\text{Mo}_2$ type

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The magnetic properties of the tetragonal ThMn_{12} -type $R\text{Co}_{10}\text{Mo}_2$ compounds have been investigated by means of magnetic measurement and x-ray diffraction. It is found that the Co moment is roughly the same in all $R\text{Co}_{10}\text{Mo}_2$ compounds ($0.9 \mu_B/\text{Co}$), excepting $\text{CeCo}_{10}\text{Mo}_2$ ($0.7 \mu_B/\text{Co}$). It is also found that the Co sublattice displays easy c -axis magnetization. Nonaxial behavior at room temperature is observed only for $\text{SmCo}_{10}\text{Mo}_2$, whereas in $\text{ErCo}_{10}\text{Mo}_2$ a spin reorientation occurs from axial to planar magnetization upon cooling below $T_{sr}=125$ K. These results suggest that the A_2^0 term, mainly responsible for the rare-earth-sublattice anisotropy has the same sign as in the $R_2\text{Fe}_{14}\text{B}$ series ($A_2^0 > 0$). This is in contrast to other ThMn_{12} -type compounds ($R\text{Fe}_{10}\text{V}_2$, $R\text{Fe}_{11}\text{Ti}$) where it is generally assumed that $A_2^0 < 0$. The intersublattice coupling of $\text{ErCo}_{10}\text{Mo}_2$ has been determined by means of the high-field free-powder method.

I. INTRODUCTION

Many novel Fe-rich rare-earth-based permanent-magnet materials have comparatively low Curie temperatures and hence require Co substitution for Curie temperature enhancement. Well-known examples are $\text{Nd}_2\text{Fe}_{14}\text{B}$, $\text{SmFe}_{11}\text{Ti}$, and $\text{SmFe}_{10}\text{Mo}_2$. In order to study the effect of Co substitution on Curie temperature, magnetization, intersublattice-coupling constant, and anisotropy field in $R\text{Fe}_{10}\text{Mo}_2$ compounds, we have studied the magnetic properties of the $R\text{Co}_{10}\text{Mo}_2$ series in pure form.

II. EXPERIMENT

The compounds of the $R\text{Co}_{10}\text{Mo}_2$ type were prepared by arc-melting stoichiometric mixtures of the metallic constituents of at least 99.9% purity. After arc melting, the polycrystalline specimens were wrapped in tantalum foil, sealed into evacuated quartz tubes, and annealed at 1000°C for 4 weeks. The samples were quickly cooled to room temperature after annealing by breaking the quartz tubes under water. Subsequently, the samples were investigated by x-ray powder diffraction and found to be approximately single phase. The x-ray patterns were indexed on the basis of the ThMn_{12} -type of structure. For each compound, the free-powder magnetization was measured at 4.2 K as a function of applied magnetic fields up to 35 T, using the High-Field Installation at the University of Amsterdam.¹ In order to investigate the presence of spin-reorientation transitions between 4.2 K and room temperature, ac-susceptibility measurements were made on several compounds in the $R\text{Co}_{10}\text{Mo}_2$ series.

III. EXPERIMENTAL RESULTS

From x-ray-diffraction experiments on magnetically aligned powders, it was derived that at room temperature the easy-magnetic direction in the $R\text{Co}_{10}\text{Mo}_2$ compounds is parallel to the c -axis. The only exception is $\text{SmCo}_{10}\text{Mo}_2$ where the easy-magnetization direction is perpendicular to the c -axis. These results confirm earlier observations by Xu and Shaheen.²

ac-susceptibility measurements show no temperature-induced spin reorientations except in $\text{ErCo}_{10}\text{Mo}_2$ and $\text{Er}_{0.8}\text{Y}_{0.2}\text{Co}_{10}\text{Mo}_2$ (see Fig. 1). The sharp cusp observed for

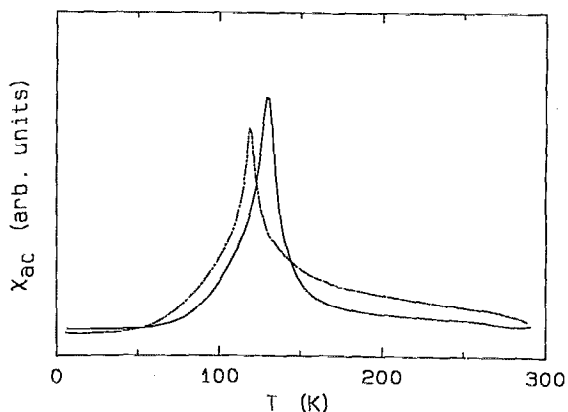


FIG. 1. Temperature dependence of the ac susceptibility for $\text{ErCo}_{10}\text{Mo}_2$ (solid line) and $\text{Er}_{0.8}\text{Y}_{0.2}\text{Co}_{10}\text{Mo}_2$ (dashed line).

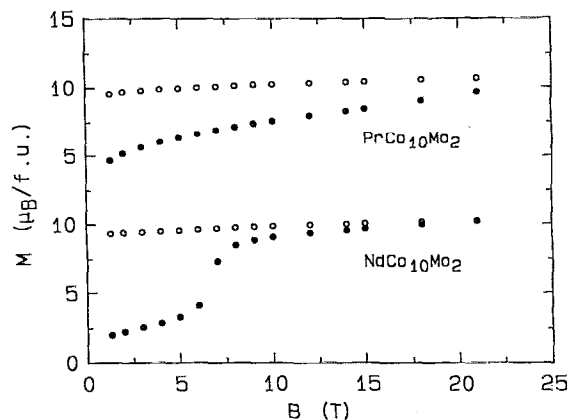


FIG. 2. Magnetic isotherms at 4.2 K measured on magnetically aligned powders of $\text{PrCo}_{10}\text{Mo}_2$ and $\text{NdCo}_{10}\text{Mo}_2$. Open circles correspond to the field parallel to the alignment direction and filled circles to perpendicular to the alignment direction.

both materials is taken as evidence of spin reorientations from $M \perp c$ (at low temperature) to $M \parallel c$ (at high temperature), at 125 K in $\text{ErCo}_{10}\text{Mo}_2$ and at 115 K in $\text{Er}_{0.8}\text{Y}_{0.2}\text{Co}_{10}\text{Mo}_2$. From the magnetic measurements on $R\text{Co}_{10}\text{Mo}_2$ compounds by Xu and Shaheen,² it can be derived that the moments of the light rare earths couple parallel to the Co moments and that the moments of the heavy rare earths couple antiparallel to the Co moments, as is usually observed.³ Therefore no information on the intersublattice-coupling strength can be derived by means of the high-field free-powder (HFFP) method^{4,5} for the compounds with the light-rare-earth elements. The isotherms of these compounds were therefore studied only in fields up to 20 T.

The magnetization at 4.2 K of magnetically aligned powders of $\text{PrCo}_{10}\text{Mo}_2$ and $\text{NdCo}_{10}\text{Mo}_2$ is shown in Fig. 2. The anisotropy field B_A for $\text{PrCo}_{10}\text{Mo}_2$ equals approximately 25 T. In $\text{NdCo}_{10}\text{Mo}_2$ there is a field-induced change of the magnetization direction, the critical field being equal to 6.5 T.

The saturation moments $M(0)$ at 4.2 K were derived from the free-powder magnetization curves by extrapolation to $B=0$. These values are listed in Table I. The anisotropy in $\text{CeCo}_{10}\text{Mo}_2$ and $\text{GdCo}_{10}\text{Mo}_2$ is predominantly that of the Co

TABLE I. Magnetic properties of $R\text{Co}_{10}\text{Mo}_2$ compounds.

Compounds	$M(0)$	M_R	M_{Co}
$\text{YCo}_{10}\text{Mo}_2$	8.5	0	8.5
$\text{CeCo}_{10}\text{Mo}_2$	6.5	0	6.5
$\text{PrCo}_{10}\text{Mo}_2$	11.2	3.2	8.0
$\text{NdCo}_{10}\text{Mo}_2$	10.6	3.2	7.4
$\text{SmCo}_{10}\text{Mo}_2$	9.5	0.7	8.8
$\text{GdCo}_{10}\text{Mo}_2$	1.9	7.0	7.0 ± 1.9
$\text{TbCo}_{10}\text{Mo}_2$	0.1	9.0	9.0 ± 0.1
$\text{DyCo}_{10}\text{Mo}_2$	0.3	10.0	10.0 ± 0.3
$\text{HoCo}_{10}\text{Mo}_2$	0.3	10.0	10.0 ± 0.3
$\text{Er}_{0.8}\text{Y}_{0.2}\text{Co}_{10}\text{Mo}_2$	1.4	7.2	7.2 ± 1.4
$\text{ErCo}_{10}\text{Mo}_2$	0.0	9.0	9.0

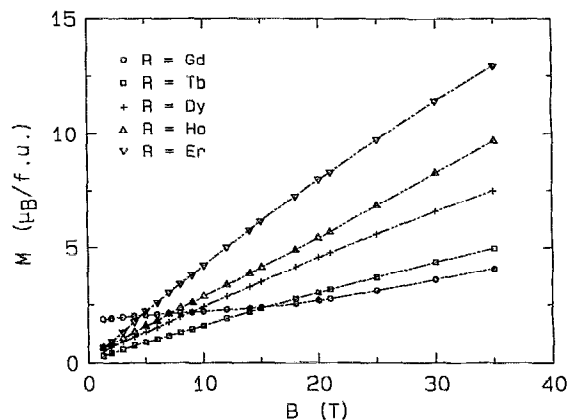


FIG. 3. Magnetic isotherms at 4.2 K of $R\text{Co}_{10}\text{Mo}_2$ compounds ($R=\text{Gd}, \text{Tb}, \text{Dy}, \text{Ho},$ and Er) measured on powder particles that are free to rotate in the sample holder.

sublattice since Ce is tetravalent and Gd is an S -state ion. The deviating valence of Ce follows from the lattice constants.

In order to obtain experimental information on the intersublattice-coupling strength we have studied how the low-field ferromagnetic state is affected by an applied field by measuring the magnetization of free single-crystalline powder particles of the compounds formed with the heavy rare earths in applied fields up to 35 T. Results obtained on the $R\text{Co}_{10}\text{Mo}_2$ compounds with $R=\text{Gd}, \text{Tb}, \text{Dy}, \text{Ho},$ and Er are shown in Fig. 3. The observation that the magnetic isotherms pass almost through the origin for the compounds with $R=\text{Tb}, \text{Dy}, \text{Ho},$ and Er means that the R -sublattice magnetization M_R and the Co-sublattice magnetization M_{Co} are nearly equal to each other in these materials.

In practically all R - T intermetallics previously investigated by means of the HFFP method,⁵ the T -sublattice anisotropy is much smaller than the R -sublattice anisotropy. In these cases, the former can be ignored and the R -sublattice magnetization can be parallel to the easy-magnetization direction and to remain so during the bending process. The free energy is given by

$$E = E_R^{an} + E_{\text{Co}}^{an} + n_{\text{RCo}} M_R M_{\text{Co}} \cos \alpha - BM. \quad (1)$$

The magnetization M is given by $M = (M_R^2 + M_{\text{Co}}^2 + 2M_R M_{\text{Co}} \cos \alpha)^{1/2}$.

In deriving the magnetization by minimizing the free energy, only the last two terms in Eq. (1) need to be taken into consideration. In this case, the intersublattice-coupling constant n_{RT} can be obtained straightforwardly from the magnetization in the region where the bending process from the ferrimagnetic configuration to the forced ferromagnetic state occurs ($n_{RT} = [M/B]^{-1}$).

In the $R\text{Co}_{10}\text{Mo}_2$ compounds, however, it is no longer legitimate to neglect the Co-sublattice anisotropy. If only the first-order anisotropy constants K_1^R and K_1^{Co} are taken into account, the free-energy expression reads

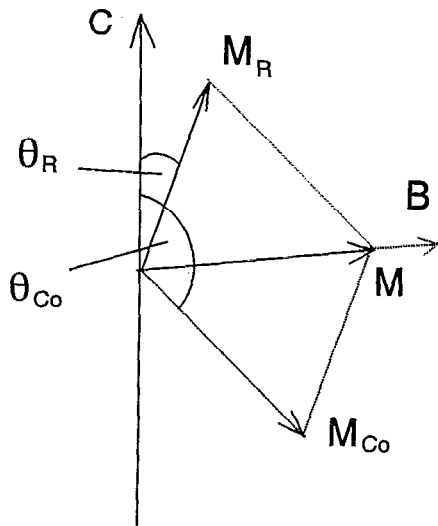


FIG. 4. Schematic representation of the orientations of M_R and M_{Co} in an external field for a free single crystal.

$$E = K_1^R \sin^2 \theta + K_1^{Co} \sin^2(\theta + \alpha) + n_{RCo} M_R M_{Co} \cos \alpha - BM. \quad (2)$$

The angles θ and α are defined in Fig. 4. Due to the presence of the two anisotropy terms in Eq. (2) neither of the two sublattice magnetizations will remain oriented along the corresponding easy-axis direction during the process that the two sublattice magnetizations bend towards each other under influence of the external field. This leads to a nonlinear behavior of the magnetic isotherm.^{10,11}

By minimizing the free energy given in Eq. (2), the following expression for the magnetization can be derived:

$$\frac{B}{M} = n_{AB} - \frac{2K_1^A K_1^B}{M_A M_B |K|} \cos \alpha, \quad (3)$$

with

$$|K| = [(K_1^A)^2 + (K_1^B)^2 + 2K_1^A K_1^B \cos 2\alpha]^{1/2}.$$

By fitting the experimental magnetization curves to this expression, details about n_{RCo} and the anisotropy constants can be obtained.

As an example we analyzed the magnetization curve of $ErCo_{10}Mo_2$. The result of the fitting procedure is shown in Fig. 5. The parameters obtained are $K_1^{Er} = -4.32 \times 10^{-22}$ J/f.u., $K_1^{Co} = 1.13 \times 10^{-22}$ J/f.u., and $n_{ErCo} = 3.06 \times 10^{23}$ T f.u./A m², $M_{Er} = 9.0 \mu_B$ /f.u., and $M_{Co} = 8.35 \mu_B$ /f.u. When expressed by means of the Hamiltonian $H = 2 J_{ErCo} S_{Er} \cdot S_{Co}$, the value of $-J_{ErCo}/k$ is equal to 6.7 K. One can see in Fig. 5 that the Er- and Co-sublattice moments will become parallel at about 60 T. In the bending process, the Er-sublattice moment oscillates around its easy direction (basal plane).

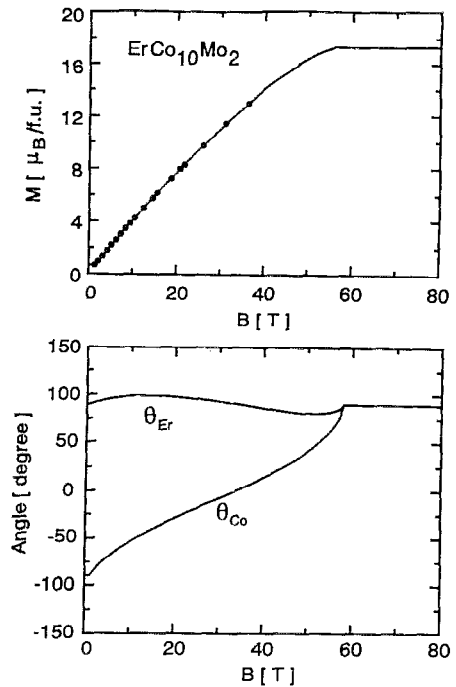


FIG. 5. (a) Comparison of the experimental and calculated magnetizations of $ErCo_{10}Mo_2$ (b) calculated field dependence of the orientations of the Er- and Co-sublattice moments.

It should be mentioned that in order to obtain the information about the magnetic coupling between R and Co sublattices for the other $RCo_{10}Mo_2$ compounds, all the magnetization curves shown in Fig. 2 should be analyzed taking into account the anisotropy of the two sublattices. This analysis which presently is carried out may be very different for the different rare-earth ions. For instance, for $R=Ho$, high-order anisotropy constants have to be taken into account.

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