# Magnetocrystalline anisotropy of novel $\mathrm{R}_{3}(\mathrm{Fe}, \mathrm{M})_{29}$ compounds 

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# Magnetocrystalline anisotropy of novel $\mathbf{R}_{\mathbf{3}}(\mathbf{F e}, \mathbf{M})_{\mathbf{2 9}}$ compounds 

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#### Abstract

The structure and magnetic properties of novel $\mathrm{R}_{3}(\mathrm{Fe}, \mathrm{M})_{29}$-type compounds ( M is the stabilizing element) have been studied by comparing to $\mathrm{CaCu}_{5^{-}}, \mathrm{R}_{2} \mathrm{Fe}_{17^{-}}$and $\mathrm{ThMn}_{12}$-type rare-earth-transition-metal compounds. A phenomenological expression of the magnetocrystalline-anisotropy energy for the monoclinic structure has been derived through symmetry analysis.


## 1. Introduction

Recently, the study of the magnetic properties of the new ternary phase with the $\mathrm{Nd}_{3}(\mathrm{Fe}, \mathrm{Ti})_{29}$-type structure is attracting considerable interest from both fundamental and practical points of view. This phase has been synthesized at the Fe-rich corner of the R (= rare-earth)-Fe phase diagram [1-3]. Initially, it has been reported that this phase has the nominal composition $\mathrm{Nd}_{2}(\mathrm{Fe}, \mathrm{Ti})_{19}$ and that the diffraction patterns can be indexed as a superlattice of the hexagonal cell of the $\mathrm{TbCu}_{7}$ type of structure. The real stoichiometric composition was later established to be 3:29. The crystallographic structure has been determined to be of the $\mathrm{Nd}_{3}(\mathrm{Fe}, \mathrm{Ti})_{29}$ type with monoclinic symmetry and $P 2_{1} / c$ space group by x-ray diffraction (XRD) and neutron powder diffraction [4-6]. Subsequent work by Kalogirou et al suggests that the $\mathrm{Nd}_{3}(\mathrm{Fe}, \mathrm{Ti})_{29}$-type structure can be described more accurately in the $A 2 / m$ space group than in the $P 2_{1} / c$ space group [7].

Like $\mathrm{R}_{2} \mathrm{Fe}_{17}$ and $\mathrm{RFe}_{12-x} \mathrm{M}_{x}$ ( M is a stabilizing element) compounds, the new family of intermetallic compounds, $\mathrm{R}_{3}(\mathrm{Fe}, \mathrm{M})_{29}$, is able to take up N and C as interstitial atoms, which leads to a remarkable improvement of the magnetic properties [8]. The interstitial nitride $\mathrm{Sm}_{3}(\mathrm{Fe}, \mathrm{Ti})_{29} \mathrm{~N}_{y}$ can be considered as a novel candidate for application in permanent magnets. Recent study on the hard magnetic properties of $\mathrm{Sm}_{3}(\mathrm{Fe}, \mathrm{M})_{29} \mathrm{~N}_{y}$ with $\mathrm{M}=\mathrm{V}, \mathrm{Cr}$ and Mo has proved the same conclusion [9-12].

The high-field magnetization and the singular point detection measurements show that Sm-based $\mathrm{R}_{3}(\mathrm{Fe}, \mathrm{M})_{29} \mathrm{~N}_{y}$ compounds exhibit a large axial magnetocrystalline anisotropy that is very important for permanent magnet application; at the same time there is an obvious magnetic anisotropy in the basal-plane for the parent compounds. This suggests that the anisotropy energy in $\mathrm{R}_{3}(\mathrm{Fe}, \mathrm{M})_{29}$ compounds with the monoclinic structure not only depends on the polar angle $\theta$, but also on the azimuthal angle $\phi$. A study of the anisotropy energy as a
function of the monoclinic structure parameters is very important, which will lead to a further understanding of the magnetocrystalline anisotropy of the novel $\mathrm{R}_{3}(\mathrm{Fe}, \mathrm{M})_{29} \mathrm{~N}_{y}$ compounds.

In this paper, the structure and magnetization properties of the $\mathrm{R}_{3}(\mathrm{Fe}, \mathrm{M})_{29}$-type compounds have been summarized in comparison with the 1:5, 1:12 and 2:17 types of structure. The magnetocrystalline anisotropy of the $\mathrm{R}_{3} \mathrm{Fe}_{29-x} \mathrm{M}_{x}$-type compounds has been studied; a phenomenological expression is presented.

## 2. Crystal structure

The $\mathrm{R}_{3} \mathrm{Fe}_{29-x} \mathrm{M}_{x}$ phase like $\mathrm{R}_{2} \mathrm{Fe}_{17}$ and $\mathrm{RFe}_{12-x} \mathrm{M}_{x}$ phases can be derived from the $\mathrm{CaCu}_{5}$ structure. Each of them represents a modification of the $\mathrm{RT}_{5}$ structure in which a fraction of the R atoms is replaced by T-metal dumbbells. This process may be described by [13]

$$
\begin{equation*}
\mathrm{R}_{1-\delta}(2 \mathrm{~T})_{\delta} \mathrm{T}_{5} \rightarrow \mathrm{RT}_{z} \tag{1}
\end{equation*}
$$

with the $2: 17$ structure corresponding to $\delta=1 / 3$ and the $1: 12$ structure corresponding to $\delta=1 / 2$. The new $3: 29$ structure corresponds to $\delta=2 / 5$. In the hexagonal $2: 17$ phase, this substitution may induce a substantial degree of disorder, leading to non-stoichiometric compounds and mixed stacking. In the rhombohedral $2: 17$ phase and the other two phases, this kind of substitution appears to be completely regular. The structure of $\mathrm{Nd}_{3}(\mathrm{Fe}, \mathrm{Ti})_{29}$ is intermediate between the well known rhombohedral $\mathrm{Th}_{2} \mathrm{Zn}_{17}$ (or hexagonal $\mathrm{Th}_{2} \mathrm{Ni}_{17}$ ) and the tetragonal $\mathrm{ThMn}_{12}$ structures, and is formed by alternating stacking of $2: 17$ and $1: 12$ segments, in the ratio $1: 1$. This can be proved by the magnetization study below.

From equation (1), it is likely that, besides the 3:29 phase, other intermediate phases based on the stacking of the rhombohedral $2: 17$ and tetragonal $1: 12$ segments may exist. For example, a 2:1 stacking ( $3 / 8$ dumb-bell replacement) would correspond to a $5: 46$ phase, whereas a 1:2 stacking ( $3 / 7$ dumb-bell replacement) would correspond to a $4: 41$ phase. According to model descriptions which are useful in the search for novel compounds [14-16], it is reasonable to suppose that many $\mathrm{R}-\mathrm{T}$ structure types are still awaiting discovery.

In the Descartes coordinate system, with $\vec{i}, \vec{j}$ and $\vec{k}$ as unit vectors along the $x, y$ and $z$ axes, the basis vectors for the unit cells of the $1: 5,2 ; 17 \mathrm{H}, 1: 12$ and $3: 29$ structures, respectively, can be written as follows [7]:
for the $1: 5$ structure

$$
\begin{align*}
\vec{a} & =\frac{\sqrt{3}}{2} a_{0} \vec{i}+\frac{1}{2} a_{0} \vec{j} \\
\vec{b} & =-\frac{\sqrt{3}}{2} a_{0} \vec{i}+\frac{1}{2} a_{0} \vec{j}  \tag{2}\\
\vec{c} & =c_{0} \vec{k}
\end{align*}
$$

for the $2: 17 \mathrm{H}$ structure

$$
\begin{align*}
\vec{a} & =\sqrt{3} a_{0}\left(\frac{1}{2} \vec{i}-\frac{\sqrt{3}}{2} \vec{j}\right) \\
\vec{b} & =\sqrt{3} a_{0}\left(\frac{1}{2} \vec{i}+\frac{\sqrt{3}}{2} \vec{j}\right)  \tag{3}\\
\vec{c} & =2 c_{0} \vec{k}
\end{align*}
$$

for the $1: 12$ structure

$$
\begin{align*}
\vec{a} & =\sqrt{3} a_{0} \vec{i} \\
\vec{b} & =2 c_{0} \vec{k}  \tag{4}\\
\vec{c} & =-a_{0} \vec{j}
\end{align*}
$$

for the 3:29 structure

$$
\begin{align*}
& \vec{a}=-2 a_{0} \vec{j}+c_{0} \vec{k} \\
& \vec{b}=\sqrt{3} a_{0} \vec{i}  \tag{5}\\
& \vec{c}=a_{0} \vec{j}+2 c_{0} \vec{k}
\end{align*}
$$

where $a_{0}$ and $c_{0}$ represent the lattice parameters of the $1: 5$ unit cell. The relations between the monoclinic $\mathrm{R}_{3} \mathrm{~T}_{29-x} \mathrm{~T}_{x}(3: 29)$ structure and the $\mathrm{RCo}_{5}$ (1:5), the hexagonal $\mathrm{R}_{2} \mathrm{~T}_{17}(2: 17 \mathrm{H})$ and the tetragonal $\mathrm{RT}_{12-x} \mathrm{M}_{x}$ (1:12) are given by the vector transformations in reciprocal space:

$$
\begin{align*}
& \left(\begin{array}{l}
h \\
k \\
l
\end{array}\right)_{3: 29}=\left(\begin{array}{ccc}
-2 & -2 & 1 \\
1 & -1 & 0 \\
1 & 1 & 2
\end{array}\right)\left(\begin{array}{l}
h \\
k \\
l
\end{array}\right)_{1: 5} \\
& \left(\begin{array}{l}
h \\
k \\
l
\end{array}\right)_{3: 29}=\left(\begin{array}{ccc}
0 & 1 / 2 & 2 \\
1 & 0 & 0 \\
0 & 1 & -1
\end{array}\right)\left(\begin{array}{l}
h \\
k \\
l
\end{array}\right)_{1: 12}  \tag{6}\\
& \left(\begin{array}{l}
h \\
k \\
l
\end{array}\right)_{3: 29}=\left(\begin{array}{ccc}
2 / 3 & -2 / 3 & 1 / 2 \\
1 & 1 & 0 \\
-1 / 3 & 1 / 3 & 1
\end{array}\right)\left(\begin{array}{l}
h \\
k \\
l
\end{array}\right)_{2: 17 H}
\end{align*}
$$

Therefore the coordinate transformation relationship of the 3:29 phase with the 1:5, 1:12 and $2: 17 \mathrm{H}$ phases is represented by:

$$
\begin{align*}
& \left(\begin{array}{l}
x \\
y \\
z
\end{array}\right)_{3: 29}=\left(\begin{array}{ccc}
1 / 5 & -1 / 5 & 1 / 5 \\
1 / 2 & -1 / 2 & 0 \\
1 / 10 & 1 / 10 & 2 / 5
\end{array}\right)\left(\begin{array}{l}
x \\
y \\
z
\end{array}\right)_{1: 5} \\
& \left(\begin{array}{l}
x \\
y \\
z
\end{array}\right)_{3: 29}=\left(\begin{array}{ccc}
0 & 2 / 5 & -2 / 5 \\
-1 & 0 & 0 \\
0 & 4 / 5 & 1 / 5
\end{array}\right)\left(\begin{array}{l}
x \\
y \\
z
\end{array}\right)_{1: 12}  \tag{7}\\
& \left(\begin{array}{l}
x \\
y \\
z \\
1
\end{array}\right)_{3: 29}=\left(\begin{array}{cccc}
-3 / 5 & 3 / 5 & 2 / 5 & -1 / 10 \\
-1 / 2 & -1 / 2 & 0 & 0 \\
3 / 10 & -3 / 10 & 4 / 5 & -1 / 5 \\
0 & 0 & 0 & 1
\end{array}\right)\left(\begin{array}{l}
x \\
y \\
z \\
1
\end{array}\right)_{2: 17 H} .
\end{align*}
$$

One can transform the atomic positions in the $1: 12$ and $2: 17 \mathrm{H}$ structures into those of the 3:29 structure. The relationships between the lattice parameters of the $3: 29$ and the $1: 5$ structure are

$$
\begin{align*}
& a=\sqrt{\left(2 a_{0}\right)^{2}+\left(c_{0}\right)^{2}} \\
& b=\sqrt{3} a_{0} \\
& c=\sqrt{\left(a_{0}\right)^{2}+\left(2 c_{0}\right)^{2}}  \tag{8}\\
& \beta=\tan ^{-1}\left(\frac{2 a_{0}}{c_{0}}\right)+\tan ^{-1}\left(\frac{a_{0}}{2 c_{0}}\right)
\end{align*}
$$

Based on the above description, the structural relation between the 3:29 and the 1:5 structure can be plotted, as shown in figure 1. The indices of the $1: 5$ structure are described as ( $h k t l$ ) with $h, k$, and $t$ representing the three axes $a_{1}, a_{2}$ and $a_{3}$ in the basal plane and $l$ representing the $z$ axis. The indices $h, k$ and $t$ obey the equation $h+k+t=0$. The $a$ and $c$ axes of the 3:29 structure are in the $\left(a_{3}, c\right)$ plane of the 1:5 structure, whereas the $b$ axis of the 3:29 structure is in the $\left(a_{1}, a_{3}\right)$ plane of the 1:5 structure. The 3:29 structure is built up from layers containing both R and Fe atoms (similar to the $z=0$ layer of the $1: 5$ structure) and layers of only Fe atoms (with the same arrangement as in the $z=1 / 2$ layer of the $1: 5$ structure) along the $[102]_{1: 5}$ direction. The main difference between the two structures concerns the mixed


Figure 1. Relations between the coordinates of the $1: 5$ and the $3: 29$ structure.

R- and Fe -atom layers. In these layers, there are two R sites, namely 2 a and 4 i . From the six R atoms around each R site in the $z=0$ layer in the $1: 5$ structure, four R atoms have been replaced by dumb-bell Fe atoms in the case of the 2 a R site, and three R atoms (in trigonal arrangement) in the case of the $4 i$ site.

## 3. Magnetocrystalline anisotropy

High-field magnetization measurements and single point detection techniques on the magnetically aligned samples showed that $\mathrm{R}_{3} \mathrm{Fe}_{29-x} \mathrm{~T}_{x}$ compounds have a comparably strong anisotropy in the plane [8]. Meanwhile, the magnetocrystalline anisotropies of the $\mathrm{Sm}_{3} \mathrm{Fe}_{29-x} \mathrm{~T}_{x} \mathrm{~N}_{y}$ nitrides are similar to that of $\mathrm{Sm}_{2} \mathrm{Fe}_{17} \mathrm{~N}_{x}$, which has a strong uniaxial anisotropy.

According to the theory presented by Birss [17], for a magnetically saturated single crystal (i.e. no domain walls in the crystal), the magnetocrystalline anisotropy energy E can formally be expressed as a series expansion:
$E=b_{i} \alpha_{i}+b_{i j} \alpha_{i} \alpha_{j}+b_{i j k} \alpha_{i} \alpha_{j} \alpha_{k}+b_{i j k l} \alpha_{i} \alpha_{j} \alpha_{k} \alpha_{l}+b_{i j k l m} \alpha_{i} \alpha_{j} \alpha_{k} \alpha_{l} \alpha_{m}+\cdots$
where $\alpha_{i}$ are the direction cosines of the magnetization relative to a rectangular Cartesian system of coordinate axes. The limitations of crystal symmetry are imposed on the tensor $b_{i j k} \ldots$ by means of equations of the form:

$$
\begin{equation*}
b_{i j k \ldots n}=\sigma_{i p} \sigma_{j q} \sigma_{k r} \ldots \sigma_{n u} b_{p q r \ldots u} \tag{10}
\end{equation*}
$$

where the $\sigma$ are the symmetry operators. If there is no preferred direction in time, which is valid as magnetocrystalline anisotropy is a static property, then all property tensors of odd rank are zero.

Normally, ten generating matrices for the 32 crystal classes can be chosen as shown below:

$$
\begin{array}{ll}
\sigma^{(0)}=[1]=\left[\begin{array}{lll}
1 & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & 1
\end{array}\right] & \sigma^{(1)}=[\overline{1}]=\left[\begin{array}{ccc}
-1 & 0 & 0 \\
0 & -1 & 0 \\
0 & 0 & -1
\end{array}\right] \\
\sigma^{(2)}=\left[2_{y}\right]=\left[\begin{array}{ccc}
-1 & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & -1
\end{array}\right] & \sigma^{(3)}=\left[2_{z}\right]=\left[\begin{array}{ccc}
-1 & 0 & 0 \\
0 & -1 & 0 \\
0 & 0 & 1
\end{array}\right] \\
\sigma^{(4)}=\left[\overline{2}_{y}\right]=\left[\begin{array}{ccc}
1 & 0 & 0 \\
0 & -1 & 0 \\
0 & 0 & 1
\end{array}\right] & \sigma^{(5)}=\left[\overline{2}_{z}\right]=\left[\begin{array}{ccc}
1 & 0 & 0 \\
0 & 1 & 0 \\
0 & 0 & -1
\end{array}\right] \\
\sigma^{(6)}=\left[3_{z}\right]=\left[\begin{array}{ccc}
-\frac{1}{2} & \frac{\sqrt{3}}{2} & 0 \\
-\frac{\sqrt{3}}{2} & -\frac{1}{2} & 0 \\
0 & 0 & 1
\end{array}\right] & \sigma^{(7)}=\left[4_{z}\right]=\left[\begin{array}{ccc}
0 & 1 & 0 \\
-1 & 0 & 0 \\
0 & 0 & 1
\end{array}\right]  \tag{11}\\
\sigma^{(8)}=\left[\overline{4}_{z}\right]=\left[\begin{array}{ccc}
0 & -1 & 0 \\
1 & 0 & 0 \\
0 & 0 & -1
\end{array}\right] & \sigma^{(9)}=\left[\begin{array}{lll}
0 & 1 & 0 \\
0 & 0 & 1 \\
1 & 0 & 0
\end{array}\right] .
\end{array}
$$

For crystals with cubic, tetragonal and hexagonal structures, the magnetocrystalline anisotropy can be described in terms of anisotropy constants [18], as summarized in table 1. For the monoclinic structure, there will be three possibilities for the point group; with generating matrix of $\sigma^{(3)}$ for the point group 2, $\sigma^{(5)}$ for the point group $m, \sigma^{(1)}$ and $\sigma^{(3)}$ for the point group $2 / m$ (see equation (11)). In the following, the forms of general polar tensors of second and fourth rank for this crystal symmetry are formulated. The non-zero components of these tensors may be written in compact form by using the notation of [17]:

$$
\begin{align*}
& b_{11}=b_{11} \\
& b_{22}=b_{22} \\
& B_{2}:{ }_{b_{33}}=b_{33}  \tag{12}\\
& b_{12}(2)=b_{12} \\
& b_{1111}=b_{1111} \\
& b_{2222}=b_{2222} \\
& b_{3333}=b_{3333} \\
& b_{1112}(4)=b_{2111} \\
& B_{4}: b_{1222}(4)=b_{2221}  \tag{13}\\
& b_{1122}(6)=b_{2211} \\
& b_{1133}(6)=b_{3311} \\
& b_{2233}(6)=b_{3322} \\
& b_{1233}(12)=b_{3312}
\end{align*}
$$

Table 1. Expressions for magnetocrystalline-anisotropy energy for various crystal systems.

| Crystal class | International symbol of symmetry class | Magnetocrystalline-anisotropy energy |
| :---: | :---: | :---: |
| Triclinic [18] | 1, $\overline{1}$ | $\begin{aligned} & K_{1} \alpha_{1}^{2}+K_{2} \alpha_{2}^{2}+K_{3} \alpha_{3}^{2}+K_{4} \alpha_{1} \alpha_{2}+K_{5} \alpha_{2} \alpha_{3} \\ & \quad+K_{6} \alpha_{1} \alpha_{3}+\cdots \end{aligned}$ |
| Monoclinic [this work] | $2, m, 2 / m$ | $\begin{aligned} & K_{1} \sin ^{2} \theta+K_{2} \sin ^{2} \theta \cos 2 \phi+K_{3} \sin ^{2} \theta \sin 2 \phi \\ & \quad+K_{4} \sin ^{4} \theta+K_{5} \sin ^{4} \theta \cos 2 \phi+K_{6} \sin ^{4} \theta \sin 2 \phi \\ & \quad+K_{7} \sin ^{4} \theta \cos 4 \phi+K_{8} \sin ^{4} \theta \sin 4 \phi \end{aligned}$ |
| Orthorhombic [18] | 222, mm2, mmm | $\begin{aligned} & \sin ^{2} \theta\left(K_{1} \cos ^{2} \phi+K_{2} \sin ^{2} \phi\right)+\sin ^{4} \theta\left(K_{3} \cos 2 \theta\right. \\ & \left.\quad+K_{4} \sin ^{2} \phi \cos ^{2} \phi+K_{s} \sin ^{4} \phi\right) \\ & \quad+\sin ^{2} \theta \cos ^{2} \theta\left(K_{6} \cos ^{2} \phi+K_{7} \sin ^{2} \phi\right) \end{aligned}$ |
| Cubic [18] | 23, m3, 432, $\overline{4} 3 m, m 3 m$ | $\begin{aligned} & K_{1}\left(\alpha_{1}^{2} \alpha_{2}^{2}+\alpha_{2}^{2} \alpha_{3}^{2}+\alpha_{1}^{2} \alpha_{3}^{2}\right) \\ & \quad+K_{2} \alpha_{1}^{2} \alpha_{2}^{2} \alpha_{3}^{2}+K_{3}\left(\alpha_{1}^{2} \alpha_{2}^{2}+\alpha_{2}^{2} \alpha_{3}^{2}+\alpha_{1}^{2} \alpha_{3}^{2}\right)^{2} \end{aligned}$ |
| Tetragonal [18] | 4, $\overline{4}, 4 / \mathrm{m}, 422,4 \mathrm{~mm}, \overline{4} 2 \mathrm{~m}, 4 / \mathrm{mmm}$ | $K_{1} \sin ^{2} \theta+K_{2} \sin ^{4} \theta+K_{3} \sin ^{6} \theta+K_{4} \sin ^{4} \theta \cos 4 \phi$ |
| Trigonal [18] | $3, \overline{3}, 32,3 m, \overline{3} m$ | $\begin{aligned} & K_{1} \sin ^{2} \theta+K_{2} \sin ^{4} \theta+K_{2}^{1} \sin ^{3} \theta \cos \theta \cos 3 \phi \\ & \quad+K_{2}^{2} \sin ^{6} \theta \cos 6 \phi+K_{3} \sin ^{6} \theta \\ & \quad+K_{3}^{1} \sin ^{3} \theta \cos ^{3} \theta \cos 3 \phi \end{aligned}$ |
| Hexagonal [18] | 6, $\overline{6}, 6 / \mathrm{m}, 622,6 \mathrm{~mm}, \overline{6} \mathrm{~m} 2,6 / \mathrm{mmm}$ | $K_{1} \sin ^{2} \theta+K_{2} \sin ^{4} \theta+K_{3} \sin ^{6} \theta+K_{4} \sin ^{6} \theta \cos 6 \phi$ |

where the notations of the type $b_{1122}(6)$ denote the six components which may be obtained from the component ' 1122 ' by unrestricted permutations of its indices. It follows directly from equation (12) that

$$
\begin{equation*}
b_{i j} \alpha_{i} \alpha_{j}=b_{11} \alpha_{1}^{2}+b_{22} \alpha_{2}^{2}+b_{33} \alpha_{3}^{2}+2 b_{12} \alpha_{1} \alpha_{2} . \tag{14}
\end{equation*}
$$

Similarly, it follows from equation (13) that

$$
\begin{align*}
b_{i j k l} \alpha_{i} \alpha_{j} \alpha_{k} \alpha_{l} & =b_{1111} \alpha_{1}^{4}+b_{2222} \alpha_{2}^{4}+b_{3333} \alpha_{3}^{4}+4 b_{1112} \alpha_{1}^{3} \alpha_{2}+4 b_{1222} \alpha_{1} \alpha_{2}^{3} \\
& +6 b_{1122} \alpha_{1}^{2} \alpha_{2}^{2}+6 b_{1133} \alpha_{1}^{2} \alpha_{3}^{2}+6 b_{2233} \alpha_{2}^{2} \alpha_{3}^{2}+12 b_{1233} \alpha_{1} \alpha_{2} \alpha_{3}^{2} . \tag{15}
\end{align*}
$$

In the above expression, the factors $2,4,6$ and 12 arise from the multiplicity implicit in the sets of relations of $B_{2}$ and $B_{4}$.

Since the three direction cosines are connected by the equation $\alpha_{1}^{2}+\alpha_{2}^{2}+\alpha_{3}^{2}=1$, equation (9) may be rewritten as

$$
\begin{align*}
E=b_{33}+b_{3333} & +\alpha_{1}^{2}\left(b_{11}+b_{33}-2 b_{3333}+6 b_{1133}\right)+\alpha_{2}^{2}\left(b_{22}-b_{33}-2 b_{3333}+6 b_{2233}\right) \\
& +\alpha_{1} \alpha_{2}\left(2 b_{12}+12 b_{1233}\right)+\alpha_{1}^{4}\left(b_{1111}+b_{3333}-6 b_{1133}\right) \\
& +\alpha_{2}^{4}\left(b_{2222}+b_{3333}-6 b_{2233}\right)+\alpha_{1}^{2} \alpha_{3}^{2}\left(2 b_{3333}+6 b_{1122}-6 b_{1133}-6 b_{2233}\right) \\
& +\alpha_{1}^{3} \alpha_{2}\left(4 b_{1112}-12 b_{1233}\right)+\alpha_{1} \alpha_{2}^{3}\left(4 b_{1222}-12 b_{1233}\right) . \tag{16}
\end{align*}
$$

If the $c$-axis is assumed to be the polar axis then the polar and azimuthal angles $\theta$ and $\phi$ are related to the direction cosines $\alpha_{i}$ by the equations

$$
\begin{align*}
& \alpha_{1}=\sin \theta \cos \phi \\
& \alpha_{2}=\sin \theta \cos \phi  \tag{17}\\
& \alpha_{3}=\cos \theta .
\end{align*}
$$

Equation (16) can be rewritten, to fourth order in the $\alpha_{i}$, in the form
$E=K_{0}+K_{1} \sin ^{2} \theta+K_{2} \sin ^{2} \theta \cos 2 \phi+K_{3} \sin ^{2} \theta \sin 2 \phi+K_{4} \sin ^{4} \theta+K_{5} \sin ^{4} \theta \cos 2 \phi$

$$
\begin{equation*}
+K_{6} \sin ^{4} \theta \sin 2 \phi+K_{7} \sin ^{4} \theta \cos 4 \phi+K_{8} \sin ^{4} \theta \sin 4 \phi \tag{18}
\end{equation*}
$$

with the anisotropy constants

$$
\begin{align*}
& K_{0}=b_{33}+b_{3333} \\
& K_{1}=\frac{1}{2} b_{11}+\frac{1}{2} b_{22}-b_{33}+b_{3333}+3 b_{1133}+3 b_{2233}-2 b_{3333} \\
& K_{2}=\frac{1}{2} b_{11}-\frac{1}{2} b_{22}+3 b_{1133}-3 b_{2233} \\
& K_{3}=b_{12}+6 b_{1233} \\
& K_{4}=\frac{3}{8} b_{1111}+\frac{3}{8} b_{2222}+b_{3333}+\frac{3}{4} b_{1122}-3 b_{1133}-3 b_{2233}  \tag{19}\\
& K_{5}=\frac{1}{2} b_{1111}-\frac{1}{2} b_{2222}-3 b_{1133}+3 b_{2233} \\
& K_{6}=b_{1112}+b_{1222}-6 b_{1233} \\
& K_{7}=\frac{1}{8} b_{1111}+\frac{1}{8} b_{2222}-\frac{3}{4} b_{1122} \\
& K_{8}=\frac{1}{2} b_{1112}-\frac{1}{2} b_{1222} .
\end{align*}
$$

Taking only the $K_{1}$ term into account in equation (18), $E$ is found to be a minimum or a maximum for [001], depending only on the sign of $K_{1}$. However, when all $\sin ^{2} \theta$ terms with $K_{1}, K_{2}$ and $K_{3}$ are taken into account, there will be eight preferential directions with a tilt angle of $\pi / 4$ with respect to the $x y$ plane. This can be seen from the following anisotropy energy expressions for three directions derived from equation (18):

$$
\begin{align*}
& E_{100}=K_{0}+K_{1}+K_{2} \\
& E_{110}=K_{0}+K_{1}+K_{3}  \tag{20}\\
& E_{001}=K_{0}
\end{align*}
$$

where the subscripts on the left-hand side refer to the direction of the magnetization.
It is easy to see that when we choose an angle $\eta=\tan ^{-1}\left(K_{2} / K_{3}\right)$ equation (18) can be changed to the form obtained by Courtois et al from the symmetry analysis [19, 20]:

$$
\begin{gather*}
E=K_{0}+K_{1} \sin ^{2} \theta+K_{2} \sqrt{K_{2}^{2}+K_{3}^{2}} \sin ^{2} \theta \cos 2(\phi+\eta)+K_{4} \sin ^{4} \theta+K_{5} \sqrt{K_{5}^{2}+K_{6}^{2}} \\
\times \sin ^{4} \theta \cos 2(\phi+\eta)+K_{7} \sqrt{K_{7}^{2}+K_{8}^{2}} \sin ^{4} \theta \cos 4(\phi+\eta)+\cdots \tag{21}
\end{gather*}
$$

The studies on the single crystal $\mathrm{Y}_{3}(\mathrm{Fe}, \mathrm{V})_{29}$ have shown that the above anisotropy energy expression for the $\mathrm{R}_{3}(\mathrm{Fe}, \mathrm{M})_{29}$-type compounds can explain the magnetization curves along $a_{1: 5}, b_{1: 5}$ and $c_{1: 5}$ axes [19,20]. More single crystals are needed for further study on the crystal structure and magnetocrystalline-anisotropy properties of the $\mathrm{R}_{3}(\mathrm{Fe}, \mathrm{M})_{29}$-type compounds.

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