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Rare-earth contribution to the magnetocrystalline anisotropy energy in $R_2\text{Fe}_{14}\text{B}$

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The zero-temperature value of the magnetocrystalline anisotropy (MCA) energy of trivalent rare-earth ions and its temperature dependence have been calculated for the whole series of the $R_2\text{Fe}_{14}\text{B}$ compounds. The computations, performed with a single-ion origin of the anisotropy and within a molecular-field approximation, reveal a large local anisotropy of the rare-earth ion, 2 orders of magnitude larger than the iron anisotropy. The crystal field acting on the rare-earth ions is of comparable magnitude as the molecular field. As a consequence, the internal ferro- and ferri-magnetic structures are very sensitive to applied external magnetic fields leading to large bending angles between the rare-earth and iron sublattice magnetizations. The calculated values for the intrinsic anisotropy constants of $\text{Dy}_2\text{Fe}_{14}\text{B}$ reproduce, within a two-sublattice model, the observed value of 20 T for the apparent anisotropy field at 4.2 K. The calculated values of the local MCA energy at zero temperature and room temperature are in good agreement with the experimentally determined data.

I. INTRODUCTION

$\text{Nd}_2\text{Fe}_{14}\text{B}$ is well known to exhibit important magnetic properties in view of its application as a permanent magnet.¹⁻⁷ Despite the large number of papers on this subject (Ref. 8 and references therein), physical understanding of the mechanisms responsible for the interaction between the magnetic moments and the origin of the crystalline electric field (CEF) in these intermetallic compounds is still rather poor. Until now some indications have been found of a single-ion origin of the magnetocrystalline anisotropy (MCA) of the $4f$ moment. The main anisotropy constant follows the sign of the first multiplicative Stevens factor α_J .² A point-charge (PC) model that only takes into account positive charges localized at the rare-earth ions reproduces the correct sign of the main crystal-field parameter B_2^0 .¹ The values of the CEF parameters provided by this model, however, seem to be largely overestimated.^{1,9} It is not a surprising result that this simple model is not applicable to the $R_2\text{Fe}_{14}\text{B}$ intermetallic compounds due to the confusing role played by Fe and probably B charges, and by conduction electrons as well.

Some compounds of the $R_2\text{Fe}_{14}\text{B}$ type show peculiar phenomena, such as a rotation of the magnetic easy direction from the basal plane to the tetragonal c axis with increasing temperatures.¹⁰⁻¹² First, however, the enormously large magnetocrystalline anisotropy energy at room temperature is the most useful magnetic property.^{3,4,13} At this temperature the hard-axis magnetization of $\text{Tb}_2\text{Fe}_{14}\text{B}$, for instance, reaches only 25% of the easy-axis magnetization at an external magnetic field as large as 15 T.³ The aim of this contribution is to make an attempt to describe the magnetic properties of these compounds. We focus our interest on the temperature dependence of the magnetocrystalline anisotropy (MCA) energy and to a novel description of the magnetization curves of the rare-earth-transition-metal intermetallic compounds.

II. MAGNETOCRYSTALLINE ANISOTROPY IN $3d-4f$ COMPOUNDS: CRYSTAL-FIELD APPROACH

In $3d-4f$ intermetallic compounds, the MCA energy is composed of two contributions coming from the rare-earth and from the iron or cobalt sublattices. The iron contribution in $R_2\text{Fe}_{14}\text{B}$ is assumed to be the same as in $\text{Y}_2\text{Fe}_{14}\text{B}$. Magnetic measurements of single-crystalline $\text{Y}_2\text{Fe}_{14}\text{B}$ provide a value of 2.0 T for the anisotropy field at 4.2 K.¹ It corresponds to a value of 0.7 MJ/m³ [= 12 K/f.u. (f.u. denotes formula unit)] for the anisotropy constant of the iron sublattice, K_1^{Fe} . The positive value reflects a preference for the tetragonal (c) axis. The MCA energy of $\text{Y}_2\text{Fe}_{14}\text{B}$ increases with increasing temperature, reaching at room temperature a 50%-larger value. The origin of such unexpected behavior is still unclear.

Recently, it was shown¹⁴ that the rare-earth contribution to the MCA energy in the $R\text{Co}_5$ compounds can be calculated from a phenomenological Hamiltonian:

$$\mathcal{H}_R = B_2^0 O_2^0 + g_R \mu_B \mathbf{J}_R \cdot \mathbf{B}_{\text{mol}}^R. \quad (1)$$

The first term represents the crystalline-electric-field Hamiltonian and the second the exchange-energy term. This approach, used for the whole $R\text{Co}_5$ series, reproduced not only the zero-temperature value of the MCA energy but also described its temperature dependence, in very good agreement with experimental data.¹⁴ Encouraged by this analysis, we are now examining the results obtained by such a simple approach for the $R_2\text{Fe}_{14}\text{B}$ intermetallic compounds. The CEF parameter B_2^0 is assumed to be dominant in a full CEF Hamiltonian. It is a well-known fact that in the tetragonal $\text{Nd}_2\text{Fe}_{14}\text{B}$ structure there are two inequivalent crystallographic sites of the rare-earth ions.^{15,16} Therefore, one could expect different local-anisotropy energies for these sites. This fact, however, is ignored here for simplicity.

It can be justified by PC model calculations of Ref. 1, which provide values for the two sites differing by only 30%. Mössbauer-spectroscopy data on $\text{Gd}_2\text{Fe}_{14}\text{B}$, analyzed by Bogé *et al.*,¹⁷ provided experimental values for the CEF parameters which differ by less than 10%. Thus, the value of B_2^0 is considered here a mean value for these two sites. The exchange-energy term is written in a molecular-field approximation as the interaction of the rare-earth magnetic moment, $g_R\mu_B J_R$, with a molecular field, $\mathbf{B}_{\text{mol}}^R$. J_R is the total angular momentum of the trivalent rare-earth ion and g_R is the Landé factor. The molecular field, $\mathbf{B}_{\text{mol}}^R$, is composed of two contributions. One originates from the surrounding iron moments. Its value has recently been evaluated.¹⁸ This field strongly dominates the second contribution \mathbf{B}_{RR}^R arising from an interaction between the rare-earth spins (Table I).

It is possible to calculate the electronic structure of a trivalent R ion under the combined action of the CEF and molecular field. It is assumed that the spin-orbit coupling is strong enough to consider only the ground state of the multiplet. Then, Hund's rules provide values for the total angular momentum J_R and the Landé factor g_R . The ground state of the Hamiltonian has been calculated for two important directions of the molecular fields: parallel and perpendicular to the tetragonal axis. It turns out that the ground state, for the internal field being parallel to the tetragonal axis, has the lowest energy for the rare-earth ions with a negative sign of the factor α_J and the highest for a positive sign of α_J . This is in one-to-one correspondence with experimental observations.² In the ground state the magnetic moment of the rare-earth ion is equal to the full trivalent moment given by Hund's rule. The local MCA energy of the R ion can be calculated as

$$E_{\text{an}}(T) = F(90^\circ, T) - F(0^\circ, T), \quad (2)$$

where

$$F(\theta, T) = -k_B T \ln \text{Tr} \exp[-\mathcal{H}_R(\theta)/k_B T], \quad (3)$$

with k_B the Boltzmann constant. At zero temperature the anisotropy energy of the $4f$ ion is simply given as the difference of the ground-state energies of the Hamiltonian (1) for the molecular field being perpendicular ($\theta=90^\circ$) and parallel ($\theta=0^\circ$) to the tetragonal axis. This energy corresponds to the stabilization energy introduced in Ref. 19. The eigenfunctions provide the value of the $4f$ moment. For the evaluation of the $4f$ -ion parameters at finite temperatures, one has to employ the Boltzmann distribution function for the excited states.

III. RESULTS AND DISCUSSION

The crystal- and molecular-field input parameters of the R^{3+} ions in the $R_2\text{Fe}_{14}\text{B}$ compounds as used in this analysis are shown in Table I. The exchange (intersublattice) molecular fields are taken from Ref. 18. The calculated rare-earth intrasublattice field \mathbf{B}_{RR}^R slightly enlarges the molecular field arising from the iron sublattice. Until now, the R - R interaction was generally assumed to be negligible compared to the interaction between rare-earth and transition-metal spins. By an inspection of Table I one can notice that the $4f$ - $4f$ interaction enlarges the molecular field experienced by a $4f$ moment by about 15% in the case of Gd and Tb. For the other elements it contributes less than 10%. The CEF parameter B_2^0 is calculated using the single-ion expression²⁰ in the form

$$B_2^0 = \alpha_J \langle r_{4f}^2 \rangle A_2^0. \quad (4)$$

α_J is the first Stevens factor and $\langle r_{4f}^2 \rangle$ is taken from the Hartree-Fock calculations of Freeman and Watson.²¹ In a PC model the parameter A_2^0 represents the lattice sum over surrounding charges and, being dependent only on the spatial charge distribution, it is assumed to be constant for the isostructural compounds. Here, a value of 0.112 K pm^{-2} has been taken. The results of our calculation are presented in Fig. 1 and Table II. The zero-temperature MCA energy of the rare-earth ions is enormously large. The Pr contribution, for instance, is 30

TABLE I. The crystalline-electric-field parameter B_2^0 of the trivalent $4f$ ions and the molecular field B_{mol}^R acting on the $4f$ magnetic moment for the $R_2\text{Fe}_{14}\text{B}$ intermetallics. The CEF parameter B_2^0 is evaluated within the single-ion approximation with $A_2^0 = +0.112 \text{ K pm}^{-2}$. The intersublattice molecular field \mathbf{B}_{ex}^R is taken from Ref. 18. The field \mathbf{B}_{RR}^R is a contribution arising from the rare-earth neighbors. ΔE_{CF} and ΔE_{MF} denote the crystal- and molecular-field overall splitting.

$4f$ ion	B_2^0 (K)	ΔE_{CF} (K)	B_{ex}^R (T)	B_{RR}^R (T)	$\mu_B B_{\text{mol}}^R$ (K)	ΔE_{MF} (K)	$\Delta E_{\text{MF}}/\Delta E_{\text{CF}}$
Ce	-21.10	384	67	1.0	45.5	195	0.5
Pr	-7.09	340	100	3.4	69.5	444	1.3
Nd	-2.00	120	150	7.9	106.1	694	5.8
Sm	+11.33	204	1000	76.4	723.1	1033	5.1
Gd	0	0	200	30.2	154.6	2165	
Tb	-2.40	259	134	17.2	101.1	1820	7.0
Dy	-1.45	244	100	10.8	74.4	1489	6.1
Ho	-0.48	93	80	6.9	58.4	1168	12.5
Er	+0.53	89	67	4.3	47.7	858	9.6
Tm	+2.03	220	57	3.5	40.7	570	2.6
Yb	+6.13	221	50	1.1	34.3	275	1.2

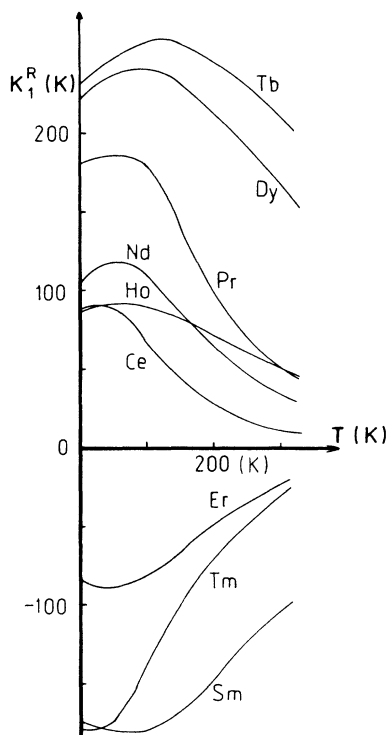


FIG. 1. The calculated temperature dependence of the rare-earth contribution to the magnetocrystalline-anisotropy energy of the $R_2Fe_{14}B$ compounds. A positive value of the anisotropy constant K_1^R indicates a preference of the rare-earth moment to the tetragonal c axis, whereas a negative value favors the easy plane. The anisotropy energy rapidly decreases with increasing temperature passing a maximum in the vicinity of 50–100 K. The Curie temperature of these compounds is of the order of 600 K.

times larger than the iron contribution to the total magnetocrystalline anisotropy. It leads to the conclusion that in the absence of an external field, the direction of the easy axis at low temperatures is solely governed by the rare-earth contribution. The largest anisotropy is attributed to Tb, Dy, Tm, and Sm ions in the $R_2Fe_{14}B$

structure. But, unfortunately, the magnetic moment of Tb, Dy, and Tm is coupled antiparallel to the magnetic moment of the $3d$ sublattice. It results in a significant reduction of the resultant magnetization. On the other hand, the Tm and Sm ions show a preference for the basal plane which makes them useless for applications. Within the series of $R_2Fe_{14}B$ compounds, the most useful properties are found for the Nd and Pr compounds. The Pr compound is expected to show even better properties than the Nd compound from an applications point of view. At room temperature the Pr anisotropy is 50% larger than the Nd anisotropy. The large zero-temperature MCA energy of the rare-earth ions rapidly decreases with increasing temperature. But even at room temperature the local anisotropy of the Nd ion is, for instance, about 30 times larger than the iron-ion anisotropy (see Table III.) The rapid decrease with increasing temperature is experimentally observed indeed.²⁴ It is interesting to notice that our computations show that the MCA energy decreases after passing through a maximum in the vicinity of 50–100 K (about a value of $0.9g_R\mu_B B_{mol}^R$). However, up until now such behavior has not been reported in experimental studies.

By an inspection of Table II one can see that the calculated value of the MCA energy at 0 K is very close to the large-exchange-limit value, $K_1^{LEL} = -3B_2^0J(J - \frac{1}{2})$. This limiting value is reached when the molecular-field effect (measured by $\Delta E_{MF} = 2g_R\mu_B J_R B_{mol}^R$) strongly dominates the crystal-field effect (measured by $\Delta E_{CF} = 3B_2^0J^2$).¹⁹ It is quite well fulfilled up to a ratio $r = \Delta E_{MF}/\Delta E_{CF}$ equal to 4. A comparison of the calculated MCA energy of $R_2Fe_{14}B$ with available experimental data is made in Table III. The total-anisotropy energy K_1^{tot} is calculated as

$$K_1^{tot} = K_1^R + K_1^{Fe} . \quad (5)$$

Quite good agreement between the calculated values of the anisotropy constant of the $R_2Fe_{14}B$ compounds and experimental data is obtained for the light-rare-earth elements. The MCA-energy values are reproduced at 0

TABLE II. The calculated zero-temperature value of the magnetocrystalline anisotropy, K_1^R , and the magnetic moment, m_R , of the trivalent $4f$ ions in the $R_2Fe_{14}B$ intermetallic compounds. The value of $K_1^{LEL} = -3B_2^0J(J - \frac{1}{2})$ is close to the calculated value in the case of the large-exchange limit, i.e., when the ratio of the molecular- and crystal-field effects r is larger than 4. The local-anisotropy field, B_A^R , and molecular field, B_{mol}^R , experienced by the rare-earth moment, are also collected.

$4f$ ion	K_1^R (K)	K_1^{LEL} (K)	r	m_R (μ_B)	B_A^R (T)	B_{mol}^R (T)
Ce	+ 90	+ 316	0.5	2.14	440	68.0
Pr	+ 179	+ 298	1.3	3.2	187	103.4
Nd	+ 105	+ 108	5.8	3.27	95	157.9
Sm	- 170	- 170	5.1	0.70	734	1076.4
Gd	0	0		7.0		230.2
Tb	+ 234	+ 238	7.0	9.0	77	151.2
Dy	+ 222	+ 228	6.1	10.0	66	110.8
Ho	+ 85	+ 86	12.5	10.0	25	86.9
Er	- 84	- 84	9.6	9.0	28	71.3
Tm	- 180	- 201	2.6	7.0	85	60.5

TABLE III. Zero- and room-temperature values of the total magnetocrystalline-anisotropy constant, K_1^{tot} , of the $R_2\text{Fe}_{14}\text{B}$ intermetallics, calculated as the sum of the rare-earth and iron contributions. The value of K_1^R represents the rare-earth contribution calculated within a single-ion approach of the anisotropy with only one CEF parameter B_2^0 by using a molecular-field approximation. The anisotropy-energy data of Ref. 2 have been calculated from the anisotropy field as determined by high-field measurements up to 35 T, whereas those of Ref. 22 have been evaluated from magnetic measurements up to 2T.

Compound	Magnetocrystalline anisotropy energy (MJ/m ³)							
	0 K				300 K			
	Calc. K_1^R	Calc. K_1^{tot}	Expt. K_1^{eff}	Ref.	Calc. K_1^R	Calc. K_1^{tot}	Expt. K_1^{eff}	Ref.
Ce	+ 10.8	+ 11.5	+ 3.4	2	+ 1.8	+ 2.8	+ 1.5 ^a	13
			+ 1.8	22				
Pr	+ 20.6	+ 21.3	+ 18.4	2	+ 6.1	+ 7.1	+ 5.4 ^a	13
			+ 24.0	22				
Nd	+ 12.3	+ 13.0	Cone		+ 4.3	+ 5.3	+ 4.8	1
			+ 21.9	2			+ 5.0	3
			+ 12.0	22			+ 4.5	4
Sm	-20.8	-20.1	-24.9	2	-12.5	-11.5	-12.0	4
			-26.0	22			-11.0	23
			-21.0	23				
Gd	0	+ 0.7	+ 0.6	17				
Tb	+ 27.6	+ 28.3	+ 6.9	22	+ 25.1	+ 26.1	+ 15 ^a	3
Dy	+ 26.6	+ 27.3	+ 4.4	2	+ 20.0	+ 21.0	+ 4.8 ^a	13
			+ 3.8	22				
Ho	+ 10.3	+ 11.0	Cone	11	+ 6.0	+ 7.0		
			+ 3.3	22				
Er	-10.2	-9.5	-7.1	2	-3.7	-2.7		
			-1.4	22				
Tm	-21.9	-21.2	-6.2	2	-4.8	-3.8		
			-3.6	22				

^aCalculated from the magnetization curves shown in Refs. 3 and 13.

K and at room temperature as well. The cerium compound is an exception. A mixed-valence state of the Ce ion is likely to be the reason for the observed smaller value. The tetravalent Ce ion will exhibit no anisotropy at all. A higher value than 3 for the valency of Ce can be deduced by comparing the lattice parameters of $\text{Ce}_2\text{Fe}_{14}\text{B}$ with those of the whole $R_2\text{Fe}_{14}\text{B}$ series. It is a well-known fact that the mixed-valence behavior is an imminent feature of cerium compounds.

Agreement between the calculated and experimental values for the anisotropy constant for the heavy-rare-earth elements is rather poor. The experimental values K_1^{eff} have been derived from a Sucksmith-Thompson analysis of the magnetization curve or/and from the apparent anisotropy field. The Sucksmith-Thompson analysis, however, fails for ferro- and ferrimagnets when the coupling between the two magnetizations is not infinitely strong, as it is in the case for the heavy rare-earth compounds, in particular. The same happens with the $R_2\text{Co}_{17}$ compounds, where magnetic measurements provide values for an effective (apparent) anisotropy energy which is at least 1 order of magnitude smaller than that derived from microscopic measurements (inelastic neutron scattering).²⁵ The values for the intrinsic anisotropy energies can be deduced from magnetization studies working within a two-sublattice model analysis.²⁶ In the case of comparable values of the anisotropy field and ex-

change field, the sublattice magnetization of the rare-earth and iron moments do not rotate as a rigidly coupled pair. Even low values of the bending angle have enormous effects on the magnetization curve.²⁷ First of all, it introduces a significant reduction of the intrinsic

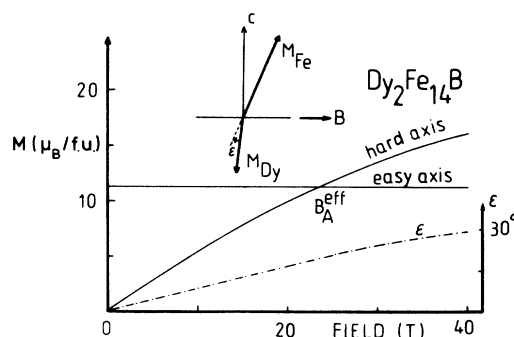


FIG. 2. The calculated magnetization curve of $\text{Dy}_2\text{Fe}_{14}\text{B}$. The calculation has been performed within a two-sublattice model with the parameters: $K_1^{\text{Dy}} = 26.6 \text{ MJ/m}^3$, $K_1^{\text{Fe}} = 0.72 \text{ MJ/m}^3$, $M_{\text{Dy}} = 100 \text{ Am}^2/\text{kg}$, and $M_{\text{Fe}} = 156.3 \text{ Am}^2/\text{kg}$. The intersublattice molecular-field coefficient $n_{\text{Dy-Fe}} [= 0.64 \text{ Tkg/Am}^2]$ (Ref. 18) is not able to assure a rigid rotation of the two (dysprosium and iron) sublattice magnetizations even at low external fields. The configuration of the two magnetization vectors at a field of 15 T (applied along the hard axis) is shown in the upper part of the figure.

MCA energies and the simple sum (5) is not valid any more. Relations between the intrinsic anisotropy constants of the two different sublattices and the effective anisotropy constants of the resultant magnetization were already derived in Ref. 28 for small values of the bending angle. Our computations indicate, however, that in realistic circumstances the bending angle can be even as large as 20° . In order to trace the effect of the bending of the two-sublattice magnetization vectors, we have calculated, within the two-sublattice model, the magnetization curve for $\text{Dy}_2\text{Fe}_{14}\text{B}$ with the parameters as given in Table I. The theoretical magnetization curve is presented in Fig. 2. The apparent anisotropy field of 23 T agrees very well with the value of 20 T obtained by Sinnema *et al.*² In a field region up to 10 T the curvature of the magnetization curve is practically invisible and the slope of this curve provides a value for the effective anisotropy constant of 4.6 MJ/m^3 . This value is in good agreement with the value of 3.8 MJ/m^3 derived by Yamauchi *et al.*²² and a value of 4.4 MJ/m^3 derived in Ref. 2. The former value was obtained by an analysis of magnetization data in an external field up to 20 kOe. It means that this bending is responsible for the 6-times-smaller value for the anisotropy constant that is observed in experimental studies as compared to the calculated intrinsic value. The reduction by the same factor is expected to be present in the other ferrimagnetic compounds as well.

IV. CONCLUSIONS

The zero-temperature value of the magnetocrystalline anisotropy energy of the trivalent rare-earth ion and its temperature dependences have been calculated for the whole series $R_2\text{Fe}_{14}\text{B}$ using only one free parameter: $A_2^0 = +0.112 \text{ K pm}^{-2}$. However, in order to explain a conical magnetic structure for $\text{Nd}_2\text{Fe}_{14}\text{B}$ and $\text{Ho}_2\text{Fe}_{14}\text{B}$, it seems to be necessary to involve higher crystal-field terms, which may become equally important at low temperatures. The crystal field acting on the rare-earth ion in these compounds is very large and of comparable magnitude to the molecular field. For Ce, Pr, and Tm ions the crystal field is even larger than the molecular field acting on the rare-earth moment. It leads to a significant reduction of the large-exchange-limit value of the MCA energy of the $4f$ ions. The highest MCA energy is attributed to Tb and Dy ions. It explains the remarkable increase of the anisotropy field of $\text{Nd}_2\text{Fe}_{14}\text{B}$ while substituting some of the neodymium atoms by dysprosium atoms.⁴ The values of the MCA energy at 0

K and at room temperature are in good agreement with the experimental data for the light-rare-earth elements. The relatively small value of the anisotropy energy that is experimentally observed for the heavy-rare-earth compounds and that is much smaller than one could expect from the single-ion mechanism is explained by a large influence of the external magnetic field on the ferrimagnetic structure. Due to its large preference for the easy axis, the rare-earth moment tilts only slightly out of the easy axis, while the increase of the magnetization for the field applied along the hard axis is mainly caused by a much easier rotation of the iron sublattice magnetization toward the external field. Magnetic measurements provide an apparent anisotropy field that must be understood as a field at which the magnetization for the field applied along the hard axis reaches the easy-axis value. A Sucksmith-Thompson analysis,²⁹ which is commonly used for the interpretation of magnetization curves, provides values for the effective anisotropy constants. These effective anisotropy constants are a complex function of the intrinsic MCA energies of the two sublattices and of the ferri- and ferromagnetic interaction between these magnetizations. The lack of any correlation between the effective anisotropy constants of the compounds with different rare-earth elements is due to this bending. An analysis of the experimental magnetization curves, within the two-sublattice model for the magnetic structure of the $3d-4f$ ferro- and ferrimagnets, permits a determination of the intrinsic anisotropy constants. The experimental observations prove that the intersublattice coupling of the $3d$ and $4f$ magnetizations is not strong enough to assure a rotation of the two sublattice magnetizations as a rigid pair even at low external fields, and that the bending effect must be taken into account for ferri- and ferromagnetic configurations of all $3d-4f$ intermetallics.

It is worth noting that spin-reorientation phenomena observed for the $R_2\text{Fe}_{14}\text{B}$ compounds with positive values of the Stevens factor α_J can be easily explained within the present model. Detailed computations are in progress and results will be published elsewhere.

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