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Canonic dependences for single-ion anisotropy and magnetostriction on magnetization

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Abstract. The magnetic anisotropy coefficients and constants of single-ion origin as functions of magnetization have been computed and classified. They are valid for a whole class of untrivial theories and are thus universal or *canonic* within this class. The role of the anisotropy *coefficients* as a finite basis of functions spanning the variation of the *constants* with magnetization and, hence, with temperature and applied field is clarified. The ratio of the zero-temperature anisotropy constants is of crucial importance for the observed type of magnetization dependence of the first anisotropy constant when two basis functions are considered. The method is directly applicable to the analysis of magnetostriction of single-ion origin and serves to identify and quantify three generic types of variation of anisotropy and magnetostriction. It is demonstrated that a compensation point $\lambda_S = 0$ for the macroscopic magnetostriction constant of amorphous ferromagnets may result from the competition between the lowest and the next-lowest single-ion contributions even if no two-ion contributions are considered. Prospective extensions of the method are given.

1. Introduction

There exist two established ways to characterize that part of the free energy of a crystalline substance which depends on the orientation of macroscopic magnetization with respect to the crystallographic axes. One way is to expand the anisotropy energy in symmetry-dictated combinations of powers of direction cosines of magnetization, whereby the description is in terms of the set of anisotropy constants $\{K_i\}$. Alternatively, one may expand in spherical harmonics and this gives rise to the description in terms of the set of anisotropy coefficients $\{\bar{\kappa}_n\}$ [1–3]. Both possibilities are of equal group-theoretical value on the phenomenological level of description of anisotropy, for the direction cosines are nothing but the reduced Descartes components of magnetization and the relationship between both descriptions is analogous to a description of a given physical property in Descartes and in spherical coordinates, respectively. Experimentally, one measures the anisotropy constants, while there is a certain theoretical advantage, even at the phenomenological level, in using the anisotropy coefficients which is due to the straightforwardness of the spherical expansion. Once the normalization of the spherical harmonics has been chosen, simple transformation relations between constants and coefficients and vice versa can be written down and these have been known for quite some time. For fixed experimental conditions, one hardly needs to know more. In an adequate thermodynamic description, however, one needs to know, beside the angular dependence, the temperature and magnetic field dependences of the anisotropy energy. In the above descriptions, they are carried by the constants $\{K_i(T, H)\}$ or the coefficients $\{\bar{\kappa}_n(T, H)\}$, respectively. In deducing the temperature and field dependences by going to the deeper level of the statistical mechanical description based on quantum mechanics, the set of coefficients is certainly the more fundamental one. The natural course of action would then be to eventually compute the coefficients as functions of temperature and field and to make use of the general, symmetry-based connection between constants and coefficients in order to find the constants as well.

In uniaxial symmetry, the anisotropy free energy F'_A in the 'constants' representation is given by

$$F'_A = \sum_{p \text{ integer}} K_p \sin^{2p}(\theta) \tag{1}$$

where θ is the angle between the direction of magnetization and the crystallographic axis of uniaxial symmetry. In the 'coefficients' representation F_A, which is equivalent to F'_A so far as the angular dependence is concerned,

$$F_A = \sum_{n \text{ even}} p_n(J) B_n^0 Y_n^0(\theta) \bar{\kappa}_n(T) \tag{2}$$

where B_n^0 are the Elliott–Stevens crystal-field parameters [4, 5], $Y_n^0(\theta)$ are the spherical harmonics, and the anisotropy coefficients are defined as the thermal averages of the Stevens operators $\langle \hat{O}_n^0 \rangle(T)$ *normalized* against their zero-temperature values [1, 3, 6]:

$$\bar{\kappa}_n(T) \equiv \frac{\langle \hat{O}_n^0 \rangle(T)}{\langle \hat{O}_n^0 \rangle(0)}. \tag{3}$$

The zero-temperature values $p_n(J) \equiv \langle \hat{O}_n^0 \rangle(0)$, with J being the angular momentum quantum number, and the anisotropy coefficients themselves can be found from the information given in [4]. The former are J-dependent products, while the latter turn out to be linear combinations of the moments $M_n \equiv \langle \hat{J}_z^n \rangle$. One may refer to [6–8] for the explicit expressions which will not be reproduced here for the sake of brevity.

2. Specifying the anisotropy coefficients for a class of untrivial theories

There is a whole class of theories [9] (mean-field theory, random-phase approximation to the Green functions approach, Callen's decoupling scheme within the Green functions technique, etc [10, 11]), for which the moments and, hence, the anisotropy coefficients can be expressed as explicit functions of the generalized effective field x of Callen and Shtrikman with the help of the explicitly known generating function for the moments in this class. The intermediate steps in this procedure can be found in [6, 7]. The overall effect is that one finds

$$\bar{\kappa}_n = \bar{\kappa}_n(x) \tag{4}$$

with an explicitly known right-hand side, while, simultaneously, the reduced magnetization per magnetic site is given by

$$m(x) \equiv M_1(x) = B_J(x) \tag{5}$$

where $B_J(x) = a \coth(ax) - b \coth(bx)$ with $a = 1 + b$ and $b = \frac{1}{2}J$ is the Brillouin function. The generalized effective field itself is to be found from

$$x = \ln(1 + 1/\Phi) \tag{6}$$

where Φ is the average number of quasiparticle excitations [9].

A *formal* solution giving the anisotropy coefficients as functions of magnetization would be achieved by inverting equation (5)

$$x = B_J^{-1}(m) = x(m) \tag{7}$$

and inserting this into equation (4) so that

$$\bar{\kappa}_n = \bar{\kappa}_n(B_J^{-1}(m)) = \bar{\kappa}_n(m(T, H)). \tag{8}$$

An *explicit* solution of the type (8) is furnished by an analytical procedure for a closed-form inversion of the Brillouin function; the explicit inverse Brillouin function was found for $J = 1$ and $J = \frac{3}{2}$ (the case with $J = \frac{1}{2}$ being a trivial one) [12, 13]. Unluckily, these values of J (without $J = \frac{1}{2}$) support the second moment $M_2 = \langle \hat{J}_z^2 \rangle$ only and, hence, only $\bar{\kappa}_2(m(T, H))$ can be found explicitly for $J = 1$ and $J = \frac{3}{2}$.

Quite recently, a very effective *parametric* solution was given [6, 7]. In fact, equations (4)–(5) as they stand represent the parametric solution with the flowing parameter x. The whole physical range of change of magnetization for any temperature and field is covered uniquely when the generalized effective field x sweeps between zero and infinity. This statement has been corroborated for the mean-field theory and for the random-phase approximation [6, 7]. Here 'uniquely' means in a one to one correspondence and this stems from the *monotonicity* of the generalized effective field with respect to temperature for fixed field and with respect to field for fixed temperature. While the property of monotonicity is rather general and is certainly valid beyond the class of theories considered, it is instructive to see it on the example of the mean-field theory. There,

$$x = \frac{cm + h}{t} \tag{9}$$

with suitably reduced temperature t, field h, and magnetization m, while c is a constant. For $h = 0$, the effective field x decreases monotonically from infinity at $T = 0$ to zero at $T = T_c$. For fixed nonzero h, the same monotonic variation of x occurs, but the zero is attained for $T \to \infty$. On the other hand, for any fixed nonzero temperature T, the effective field increases monotonically with the growth of applied field h and tends to infinity for $h \to \infty$.

3. Canonic dependences and the parametric method for their computation

It must now be obvious that one does not need to specify x from equation (6) in order to find the dependence of the anisotropy coefficients $\bar{\kappa}_n$ on the magnetization. One needs only input the value of J and collect pairs of points for a given value of $x \in [0, \infty)$ in a parametric sweep with equations (4)–(5). This means that the dependences $\bar{\kappa}_n(m)$ so calculated are valid for the whole class of theories mentioned above. In this sense, they are *canonic* dependences. Until recently, only the canonic curves for $\bar{\kappa}_2(m)$, $\bar{\kappa}_4(m)$, and $\bar{\kappa}_6(m)$ with $J = \infty$ have been known. Their extensive use [1, 3, 14] is based on the fact that a closed-form expression in terms of the (suitably normalized) modified spherical Bessel functions $\hat{I}_{l+1/2}(\mathcal{L}^{-1}(m))$ with $l = 2, 4, 6, \ldots$ has been found [15], where $\mathcal{L}^{-1}(y)$ is the inverse Langevin function. Taken at face value [16], however, these functions are nothing more than combinations involving hyperbolic functions of the type arising when calculating the $\bar{\kappa}_n$ with the help of the generating function of Callen and Shtrikman [9] for *any* finite J. Besides, a closed-form expression for the inverse Langevin function is unknown just as B_S^{-1} for $J > \frac{3}{2}$. Hence, the infinite-J case is no less formal than any $J > \frac{3}{2}$ case. The parametric solution makes this difference between finite and infinite values of J ignorable, for it operates equally well and easily with *any* J, be it finite or infinite. Apart from the seeming simplicity of the infinite-J result, the point was made that the $\bar{\kappa}_n$ with finite J, when given as functions of magnetization, are practically the same as for the infinite-J case. The statement [1] was based on comparison with dependences found by Wolf [17, 18], of which, however, only $\bar{\kappa}_4(m)$ for the two values $\frac{5}{2}$ and $\frac{7}{2}$ of the spin quantum number could be an object of comparison as only their analogues for the case considered by Wolf had been given [17, 18]. However, his calculations concerning local uniaxial distortions in cubic symmetry were set up within a framework of computing quantum mechanically nonequidistant energy levels and are perturbative and restricted to sufficiently small uniaxial couplings and to low temperatures. Besides, two further objections can be raised. First, using automatically the infinite-J results implies, e.g., that one could set up a calculation in which a system of magnetic moments of, say, $J = 1$ is found to possess anisotropy coefficients of order four, six, etc. However, as is well known, higher-order anisotropies are supported only by sufficiently high values of J. A further objection is the fact that the normalizing factors $p_n(J)$, i.e. the zero-temperature values of the corresponding Stevens operators, are quite different for different values of J, and much more so for $J \to \infty$. This point has been brought to light in [19].

3.1. *Canonic dependences for the anisotropy coefficients*

All these remarks serve to clarify that the situation with the canonic dependences is rather simple for any value of J once the point of view of the parametric approach is grasped and accepted as a working tool. Its output is easy to generate and implement. In figure 1 we give the first group of canonic dependences, those for the anisotropy coefficients for different values of J. *They are valid for the whole Callen and Shtrikman class of theories* [9]. Besides, in the normalized form of the definition (3) they do not depend in any way on the normalization of the spherical harmonics or the Stevens operators [20]. The conjecture that the curves with different values of J lie close to each other is borne out. Note, however, that very serious deviations between finite and infinite values of J occur when the *temperature dependence* of anisotropy is computed theoretically. For instance, it has been proven within the mean-field approximation that $\bar{\kappa}_2(T)$ is a strictly linear function of temperature for $J \to \infty$ in zero applied field ($\bar{\kappa}_2(T) = 1 - T/T_c$), while this is by far not the case for $\bar{\kappa}_2(T)$ for any finite J [6].

3.2. *Canonic dependences for the anisotropy constants*

The second group of canonic dependences is related to the anisotropy constants as functions of magnetization. The issues involved will be discussed in sufficient detail for the uniaxial case under the assumption of negligible in-plane anisotropy, while only the salient results will be mentioned for the case of cubic symmetry. It has often been remarked that the 'unusual', non-monotonic dependences of the anisotropy constants of great many materials are presumably due to the effect of higher-order anisotropy. This issue requires a closer examination. The best way to proceed is to consider the set of anisotropy constants as expressed in terms of the set of the anisotropy coefficients. Under the assumptions of the fundamental theory of magnetic anisotropy and magnetostriction [1, 21, 22] and throughout the Callen and Shtrikman class of theories [9],

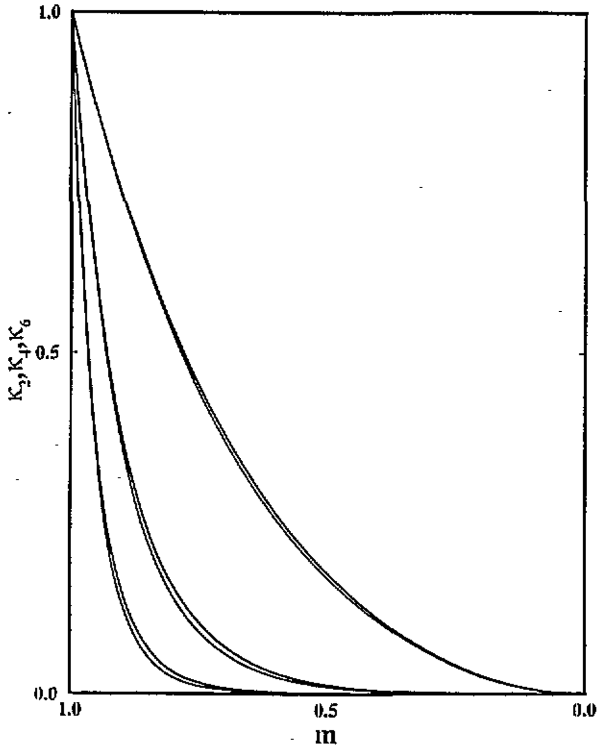

Figure 1. Canonic dependences for the normalized anisotropy coefficients: the set $\bar{\kappa}_2, \bar{\kappa}_4$, and $\bar{\kappa}_6$ as functions of magnetization (arranged in this order from above). In each family, the upper curve is for $J = \frac{7}{2}$, the lower one is for $J = \frac{15}{2}$.

the following triangular set of relations holds [8]:

$$\begin{array}{lllllllllll} K_p & = & & & & & & & & & a_p\bar{\kappa}_{2p} \\ K_{p-1} & = & & & & & & & b_{p-1}\bar{\kappa}_{2(p-1)} & + & b_p\bar{\kappa}_{2p} \\ \vdots & & & & & & & & & & \\ K_2 & = & & & c_1\bar{\kappa}_4 & + & \dots & + & c_{p-1}\bar{\kappa}_{2(p-1)} & + & c_p\bar{\kappa}_{2p} \\ K_1 & = & d_1\bar{\kappa}_2 & + & d_2\bar{\kappa}_4 & + & \dots & + & d_{p-1}\bar{\kappa}_{2(p-1)} & + & d_p\bar{\kappa}_{2p}. \end{array} \tag{10}$$

An extensive discussion about what is the highest anisotropy constant K_p which appears and, hence, what is the number p of these relations under various circumstances has been given elsewhere [8]. In any case, for single-ion anisotropy due to rare earth atoms (and under the assumptions just mentioned), there is no higher constant than K_3 [5, 23]. Once the number of anisotropy constants relevant to the discussion in any given material is settled, the triangular property of the above relations is seen to come from the fact that a term of the form $K_i \sin^{2i}\theta$ in the phenomenological expansion cannot contain contributions from $\bar{\kappa}_{2q} Y_q^0(\theta)$ with q smaller than i, since this would imply that K_i multiplies also terms of lower powers in $\sin\theta$ which is impossible by the definition of K_i. The very important point we wish to emphasize is that the set of normalized, dimensionless, monotonic functions $\{\bar{\kappa}_{2p}\}$, defined on the interval [0, 1] and taking on values between [0, 1], sets up a *basis of functions* which carry the dependence on magnetization m and, via m, on temperature and applied field as well. Hence, the relations (10) should be regarded not so much as expressing the otherwise correct fact of mutual interchangeability of *constants* and *coefficients* when it comes to the characterization of anisotropy in a given system, but as relations expressing a set of unknown functions (the K_i^0) as linear combinations of a finite basis of functions (the $\bar{\kappa}_n$) which can be easily computed for a whole class of untrivial theories by implementing the parametric method as explained earlier in this paper. Now then 'higher-order anisotropy' means that one must consider $p > 1$ constants in the phenomenological expansion. The 'must' may originate from the theoretical side (for some firmly established reasons, one is set from the beginning to consider a basis of p functions $\bar{\kappa}_n$) or, more reasonably, it may derive from practical considerations. One namely considers the top relation in (10) and notes that, since $\bar{\kappa}_{2p}$ is dimensionless and normalized so as to be unity at $T = 0$, the coefficient a_p is identical with $K_p^0 = K_p(T = 0)$. If there is experimental information that the latter quantity is small beyond concern, one may cut down the set of constants by one. In other words, arguments of smallness *as applied to the intrinsic* ($T = 0$) *anisotropy* may happen to be applicable. Once the number p of constants to be considered has been decided upon, one notes that the coefficients in the linear combinations (2) are expressible via p independent quantities. It seems especially useful and promising to take the zero-temperature values $\{K_p^0\}$ as p independent quantities [8]. The number can be reduced by one to $p - 1$ by expressing all anisotropy constants in units of any one of them and it is natural to choose the unit as K_1^0 (care has to be exercised in tracking down the effect of its *sign*, however).

4. Types of canonic curve in the two-constant case

To illustrate these ideas, we choose the simplest case involving higher-order anisotropy, the one with $p = 2$. In the following, we find it convenient to use the normalization for the spherical harmonics as given in [1] and [2]. This is important if connection is to be established with *ab initio* calculations where, typically, the crystal field parameters $\{A_n^m\}$ entering the $\{B_n^m\}$ are being calculated. Note, however, that neither the product $p_n(J)B_n^0Y_n^0$ entering equation (2) nor our results concerning the anisotropy constants depend on the normalization chosen. The relevant relations in the two-parameter ($p = 2$) case are, in units of K_1^0,

$$\frac{K_1}{K_1^0} = \left(1 + \frac{8}{7}r\right)\bar{\kappa}_2 - \frac{8}{7}r\bar{\kappa}_4 \qquad (11)$$

$$\frac{K_2}{K_1^0} = r\bar{\kappa}_4 \qquad (12)$$

where we have introduced the ratio $r \equiv K_2^0/K_1^0$. Note that the use of a two-parameter representation of anisotropy is in fact exact for quantum numbers $J = 2$ and $J = 5/2$, while for cases with $J > 5/2$ it might still be a reasonable approximation. This will be the case, for instance, with single-ion anisotropy coming from rare earth atoms with $J > 5/2$ in materials for which K_3^0 is negligibly small [8].

To summarize the information on the second group of canonic curves, one generates the dependence of the anisotropy *constants* on magnetization by implementing the parametric method to equations (11) with insertion of the relevant value of the ratio r. This means that, so far as the constants in the two-constant approximation are concerned, one has to do with a one-parameter family of canonic curves, unlike the totally determined basis functions $\bar{\kappa}_n(m)$. Still, for a given r, the curves are valid for the whole class of theories as defined earlier. In figure 2(a, b) we give two such families of curves for $J = \frac{7}{2}$ and $J = \frac{9}{2}$, respectively. In the latter case, the curve with $r = -3$ corresponds tentatively to the experimental ratio K_2^0/K_1^0 measured in $Fe_2Nd_{12}B$. Only the dependences $K_1(m)$ are presented, as by equation (11)

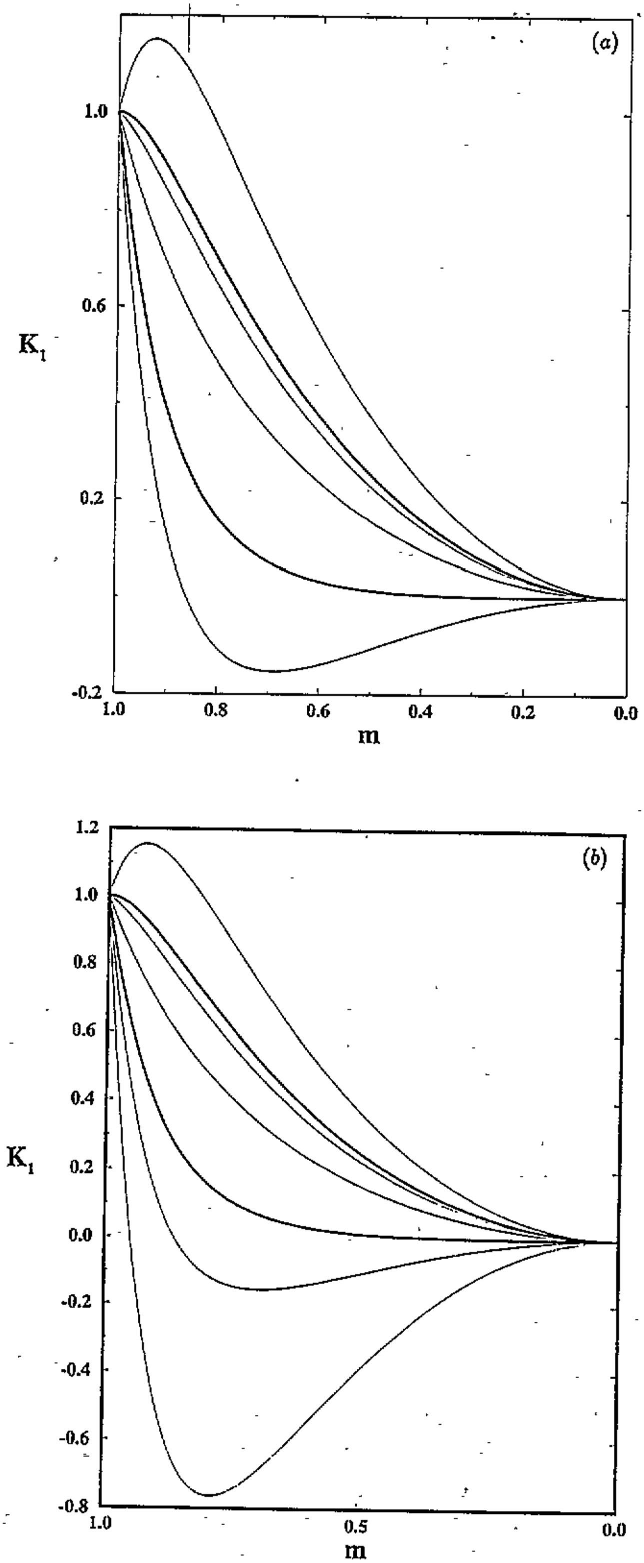

Figure 2. Canonic dependences for the anisotropy constant $K_1(m)/K_1^0$ in the two-constant uniaxial case for a given intrinsic ratio $r \equiv K_2^0/K_1^0$. The thick curves are the borderlines between the three generic types of dependence and are obtained with $r = \frac{3}{8}$ and $r = -\frac{7}{8}$ (upper and lower thick curves, respectively). Representative values of r have been taken to illustrate the dependence inside each of the generic groups of behaviour (thin lines). (a) $J = \frac{7}{2}$: from above $r = 1, \frac{1}{4}, -\frac{1}{8}, -\frac{3}{2}$; (b) $J = \frac{9}{2}$: from above $r = 1, \frac{1}{4}, -\frac{1}{8}, -\frac{3}{2}, -3$.

the dependence $K_2(m)$ is given by $\bar{\kappa}_2(m)$ up to scale and sign which are dictated by r. One can see that the expectation for a richer variety of dependences for the constants $K_i(m)$, as opposed to the 'cool' monotonicity of the coefficients $\bar{\kappa}_n(m)$ (figure 1), has been met. Three types of dependence are easily distinguished in figure 2; they are separated by thick borderlines in the same figure. The origin of the variety observed for $K_1(m)$ can also be easily understood. This is simply the extra basis function $\bar{\kappa}_2(m)$ contributing to $K_1(m)$, but not to $K_2(m)$. Two general features will certainly persist for cases with more constants included. First, in principle, one will always observe the most peculiar behaviour with the anisotropy constant of lowest order (K_1), since $K_1(m)$ is always the result of superposing the greatest number of basis functions $\bar{\kappa}_n(m)$, except for some ratios which nullify some factor in the linear combination (see below for an example). Second, for sufficiently high temperatures (sufficiently small magnetization) it is the first anisotropy constant that will dominate, regardless of what the $p-1$ ratios $r_j = K_j(T=0)/K_1(T=0)$ $(j = 2, 3, \ldots, p)$ are. This is easily derivable from the asymptotic expansions of $\bar{\kappa}_n(m)$ for $T \to T_c$ $(m \to 0)$ [8] and can be seen in the canonic plots in figure 1.

Consider now the types of curve for $K_1(m)/K_1^0$ given in figure 2. Generically, they are of (i) a non-monotonic type with a maximum (above the upper borderline), (ii) a *monotonic* type (in the domain between the borderlines), (iii) a non-monotonic type with a negative minimum and a zero point at some inner point m_S (below the lower borderline). Mathematically, the first and third types are equally peculiar. They both have a local extremum at an internal point of the interval $m \in [0, 1]$. Physically, the third type is much more intriguing, since it involves a change of sign of K_1. Imposing the condition $K_1 = 0$, one gets

$$\frac{8}{7}\frac{r}{1+\frac{8}{7}r} = \frac{\bar{\kappa}_2(m_S)}{\bar{\kappa}_4(m_S)} > 0. \tag{13}$$

The inequality could be met for either $r > 0$ or $r < -\frac{7}{8}$. The first possibility ($r > 0$) is ruled out by counterexample (take, e.g., $r = 1 > 0$, substitute in equation (11), and mind that $\bar{\kappa}_2$ is strictly greater than $\bar{\kappa}_4$ everywhere inside $m \in (0, 1)$ (figure 1)). So the lower borderline corresponds to a critical value of $r = -\frac{7}{8}$.

This simple argument has the deficiency to be a necessary condition only and the sufficiency is in fact proven by the parametric method (cf. figure 2). However, an exhaustive argument leading to the analytical determination of both borderlines can be set up for the two-constant anisotropy case we are now considering. It is based on examining the signs of the first derivatives of the basis functions $\bar{\kappa}_n$ at the ends of the interval $m \in [0, 1]$ with the help of the explicit asymptotic expressions for these functions. The method has been employed in a study of the *temperature* dependence of the anisotropy constants in the mean-field approximation and has been given in detail elsewhere [8]. One finds that the family of canonic curves for $K_1(m)$ splits into three generic groups specified as follows:

$$\text{group 1:} \quad r > \tfrac{3}{8} \tag{14}$$

$$\text{group 2:} \quad -\tfrac{7}{8} \leqslant r \leqslant \tfrac{3}{8} \tag{15}$$

$$\text{group 3:} \quad r < -\tfrac{7}{8}. \tag{16}$$

The borderlines are thus to be found for $r = \frac{3}{8}$ and $r = -\frac{7}{8}$. By equation (11), at $r = \frac{3}{8}$, $K_1(m)/K_1^0 = \frac{10}{7}\bar{\kappa}_2(m) - \frac{3}{7}\bar{\kappa}_4(m)$, while at $r = -\frac{7}{8}$ one finds simply $K_1(m)/K_1^0 = \bar{\kappa}_4(m)$ and $K_2(m)/K_1^0 = -\frac{7}{8}\bar{\kappa}_4(m)$, i.e., $K_2(m) \sim K_1(m) \sim \bar{\kappa}_4(m)$. In this latter case, we have an example of a particular value of the ratio r for which both linear superpositions (cf. equation (2) for K_1 and K_2 are of 'equal length' (here, simply proportional to $\bar{\kappa}_4$, while $\bar{\kappa}_2$ does not show up at all). Accordingly, the lower borderline in figure 2 is just the plot of the basis function $\bar{\kappa}_4(m)$.

The classification of the possible types of dependence of K_1 as a function of *magnetization* is the same as found in the analysis of the corresponding *temperature* behaviour in the mean-field approximation. As it stands here, this classification is valid for the whole class of theories envisaged [9], since here it is found when considering the relevant basis functions as functions of *magnetization* and it was already demonstrated above that these are valid for the whole class. This result is in itself a generalization in comparison with the mean-field treatment of the *temperature* dependence [8]. Moreover, the temperature behaviour of the first anisotropy constant in the two-constant anisotropy case will definitely obey the classification not only for the mean-field case, but for the whole Callen and Shtrikman class. This is essentially due to the monotonic behaviour of magnetization with temperature and it is the invariance of the sign of $dm/dT < 0$ that matters when extending the above classification to any particular temperature dependence within the class.

For the sake of completeness, we mention in passing that the same type of analysis leads in the two-constant cubic case to the subdivision of the family of canonic curves for $K_1^{cubic}(m)$ into three analogous generic types, which are realized for $r > 10$, $-11 < r < 10$, and $r < -11$, respectively. One sees that there is a substantial quantitative difference by an order of magnitude, so far as the values of r delineating the analogous regimes are concerned. This makes the possibility for an experimental realization of the non-monotonic types of behaviour rather unrealistic. Besides, in cubic materials for any given anisotropy constant K_j^{cubic} the linear combinations are one term shorter and, consequently, the variation opportunities are significantly reduced. Note, however, that if one considers anisotropy of single-ion origin due to rare earth ions sitting on sites of cubic symmetry, the classification presented here for the cubic case is *exhaustive*, since no basis functions $\bar{\kappa}_n(m)$ with $n > 6$ are allowed [23], while n must be greater than two by cubic symmetry; hence, one is left with just two basis functions, those with indices four and six.

5. Extension of the method to magnetostriction of single-ion origin

The systematization of the variation of anisotropy constants with magnetization m is immediately applicable to the analysis of the variation of *magnetostriction* of single-ion origin with magnetization. The detailed expressions for the magnetostriction coefficients are rather complicated both in uniaxial and in cubic symmetry [1, 21, 22], since group theoretical classification arguments are involved and quite a number of magnetoelastic coupling constants as well as elastic constants have to be taken into consideration. However, there is almost no hope that all this detailed information could be available with any reliable accuracy in any given experimental case. That is why one resorts to folding all unknown information into free fitting parameters which are then determined from the available data on the variation of magnetostriction with temperature or magnetization. When this course of action is adopted, the problem is seen to be immediately reduced to the characterization of the functions $\{\lambda_i\}$ (the independent components of the magnetostriction tensor) as linear combinations of several other functions carrying the magnetization dependence. To be specific, we shall not examine at this time contributions to magnetostriction beyond the single-ion ones; furthermore, the latter will be treated within the Callen and Callen theory [1] which is valid for the whole Callen and Shtrikman class [9]. To be able to apply the results from the analysis of magnetic anisotropy as given above, we keep the contributions from $\bar{\kappa}_2(m)$ and $\bar{\kappa}_4(m)$ or from $\bar{\kappa}_4(m)$ and $\bar{\kappa}_6(m)$ in the case of uniaxial or cubic symmetry, respectively. All this amounts, in the first place, to writing the independent components $\lambda_i(m)$ as

$$\lambda_i(m) = c_1^i \bar{\kappa}_{2z}(m) + c_2^i \bar{\kappa}_{2(z+1)}(m) \tag{17}$$

where c_1^i and c_2^i incorporate the unknown detailed information about the magnitude of the magnetoelastic coupling and the elastic constants involved, but do not depend on magnetization, while the number z appearing in the subscripts of the basis functions is one or two for the uniaxial or cubic cases, respectively. Now that the basis of functions has been set up, it is not essential that the coefficients in the latter linear combination have not some deeper meaning like the meaning of transformation coefficients between equivalent representations in the case of magnetic anisotropy which we considered first. Using normalization against the zero-temperature value of magnetostriction in order to reduce the number of coefficients by one as before, one finds

$$\frac{\lambda_i(m)}{\lambda_i(m=1)} = \alpha^i \bar{\kappa}_{2z}(m) + (1-\alpha^i)\bar{\kappa}_{2(z+1)}(m) \tag{18}$$

where use was made of the normalization of the $\bar{\kappa}_n$ ($\bar{\kappa}_n = 1$ for $m = 1$, i.e., for $T = 0$). The conclusions about the possible types of behaviour of the respective magnetostrictive constant due to single-ion anisotropy derive immediately from the analysis, already done for the anisotropy constants. Namely, three types of variation with magnetization (and, hence, with temperature) are possible and these can be defined for the whole class of theories under consideration.

Now one finds easily the domains of the three generic groups of canonic curves for magnetostriction. In uniaxial symmetry, one finds

$$\text{group 1:} \quad \alpha^i > 10/7 \tag{19}$$

$$\text{group 2:} \quad 0 \leqslant \alpha^i \leqslant 10/7 \tag{20}$$

$$\text{group 3:} \quad \alpha^i < 0. \tag{21}$$

The canonic curves for the three types look like those for the anisotropy constants in the three corresponding classes (cf. figure 2). The 'family parameter' for each component λ_i is now α^i and the formal correspondence with anisotropy is established via the relation $\alpha^i = 1 + \frac{8}{7}r$.

In cubic symmetry,

$$\text{group 1:} \quad \alpha^i > \tfrac{21}{11} \tag{22}$$

$$\text{group 2:} \quad 0 \leqslant \alpha^i \leqslant \tfrac{21}{11} \tag{23}$$

$$\text{group 3:} \quad \alpha^i < 0. \tag{24}$$

The correspondence with the treatment of anisotropy is established via $\alpha^i = 1 + \frac{1}{11}r$.

In both cases of uniaxial and cubic symmetry the lower borderline (the one between the monotonic type of variation and the type with a change of sign) occurs with $\alpha^i = 0$, whereby it is plain to see that this borderline is simply either $\bar{\kappa}_4(m)$ in the uniaxial case or $\bar{\kappa}_6(m)$ in the cubic case. Note that, just like with the anisotropy, the borderlines between the different generic regimes of variation *do not depend on the value of J*, although the basis functions do.

There are many ways in which the above information may be used in the analysis of magnetostriction. By way of example from the experimental side, provided that only the *type* of variation of magnetostriction of single-ion origin has been determined and even without attempting a one-parameter fit to determine the value of α^i, one may use the relevant inequality from among equations (19–21) or equations (22–24) as an integral (overall) restriction on the combinations of magnetoelastic and elastic coefficients which enter the parameter α^i explicitly. On the other hand, due to the smallness of the magnetoelastic coefficients *and* the existence of fundamental restrictions on the values of the elastic coefficients stemming from requirements of thermodynamic elastic stability [24], it may turn out to be possible to rule out some of the three predicted types or to introduce additional restrictions under which a given type may exist.

There is, however, a much more important issue of major practical interest which is involved in the analysis of magnetostriction of single-ion origin. It concerns the possibility for, and the explanation of the origin of, near-zero saturation magnetostriction $\lambda_S \approx 0$ in a series of technologically important soft amorphous Co-rich or Fe-rich ferromagnetic alloys [22, 25–28]. There arose the question of whether systems of this type are really nonmagnetostrictive on a local scale or whether they exhibit a local magnetostrictive effect which averages out on a macroscopic scale. These *structure-related* possibilities were denoted as type 1 and type 2 magnetostrictive alloys. To solve these issues, a polycrystalline model of amorphous ferromagnets has been developed [27–30] whereby it is assumed that the amorphous alloys are composed of very small structural units ('grains') which exhibit similar symmetry and chemistry, but are randomly oriented. In a locally assigned frame of reference adapted to the local symmetry exhibited, the magnetostrictive behaviour of an isolated grain may be described by the independent components $\{\lambda_i\}$ of the magnetostriction tensor, and it is assumed that the temperature dependence of λ_i in these grains is determined by two functions, each of which depends monotonically on m:

$$\lambda_i = a_i f(m) + b_i g(m). \tag{25}$$

Taking into account the elastic interactions between the distinct grains, it has been shown [28–30] that the macroscopic magnetostriction constant λ_S of the alloy may be represented as a linear combination of the $\{\lambda_i(m)\}$, i.e.,

$$\lambda_S(m) \equiv L\,[\lambda_i(m)] = L[a_i]f(m) + L[b_i]g(m). \tag{26}$$

A system with $\lambda_S\,(m(T_0)) = 0$ at some temperature T_0 may then arise if all $\lambda_i\,(m(T_0))$ are zero (type 1) or if the linear combination $L\,[\lambda_i(m(T_0))]$ is zero (type 2) and it has been argued that type 1 materials are unlikely to occur. So far, it has always been assumed in the literature that the two relevant basis functions $f(m)$ and $g(m)$ are those describing the lowest single-ion contribution and the two-ion contribution to magnetic anisotropy [22, 25, 26], i.e., that $\lambda_S = 0$ comes about as a result of the competition between two rather different sources of magnetic anisotropy and, hence, magnetostriction. In contrast, one can also assume that the coefficients a_i and b_i in equation (25) are given by the coefficients c_1^i and c_2^i of equation (17), i.e., the two relevant basis functions are related to the lowest and second-lowest single-ion anisotropy. It has been demonstrated in this section that a change of sign of $\lambda_i(m)$ may indeed be obtained if the related parameters α_i from equation (18) are within a certain range. One can then obtain a zero-magnetostrictive alloy of type 1 if *all* λ_i vanish simultaneously for some particular value of magnetization and this is very unlikely. On the other hand, the macroscopic magnetostriction constant $\lambda_S(m)$ is given by

$$\lambda_S(m) = L[c_1^i]\bar{\kappa}_{2z}(m) + L[c_2^i]\bar{\kappa}_{2(z+1)}(m) \tag{27}$$

and, hence,

$$\frac{\lambda_S(m)}{\lambda_S(m=1)} = \alpha\bar{\kappa}_{2z}(m) + (1-\alpha)\bar{\kappa}_{2(z+1)}(m). \tag{28}$$

A zero-magnetostrictive alloy of type 2 is now seen to result whenever the parameter

$$\alpha \equiv \frac{L[c_1^i]}{L[c_1^i] + L[c_2^i]} \tag{29}$$

falls into the range specified as group 3 in equations (21) and (24) for uniaxial and cubic local symmetry, respectively. We have thus demonstrated that a vanishing macroscopic magnetostriction in an amorphous alloy may arise from the competition between single-ion anisotropies of different orders, even if there is no two-ion contribution. In certain cases, it might be important for the analysis of experimental data to consider simultaneously both sources of compensation effects, namely, higher-order single-ion anisotropy and two-ion contributions (see also the end of the next section).

6. On the canonic dependences for anisotropy constants in higher-order ($p > 2$) cases

There are certainly further improvements which could be done to elaborate this type of rather general analysis. Two of these may be mentioned here as being in the phase of active exploration.

The first one is the extension of the analysis of single-ion anisotropy to characterize exhaustively the effect of extending the basis of functions $\bar{\kappa}_n$ by one for the uniaxial case. The possible types of canonic dependence for $K_1(m)/K_1^0$ will certainly be more numerous as now the corresponding linear combination is one function longer and, more importantly, depends on an additional parameter K_3^0/K_1^0. That is to say, now a two-parametric manifold has to be explored systematically to set up a full-scale classification of the kind proposed above. Besides, with a basis of three functions $\bar{\kappa}_n$ one must expect interesting behaviour for the *second* anisotropy constant $K_2(m)$ as well. Moreover, in view of the present analysis, the variation of $K_2(m)$ in this more involved case *may already now be considered as exhaustively characterized.* Indeed, in the three-parameter case $K_2(m)/K_1^0 = d_1\bar{\kappa}_4(m) + (1 - d_1)\bar{\kappa}_6(m)$, where d_1 depends on a single ratio only [2]. Using the above method and the explicit formulae for transforming between constants and coefficients in the three-parameter case, one can easily classify the values of the relevant ratio for which *three* (!) different types of behaviour of K_2 occur. These rather general predictions certainly extend the understanding as to what might be expected from the variation of the *second* anisotropy constant in uniaxial materials whose anisotropy is largely due to single-ion contributions coming from rare earth ions, for instance. It might be expected that indeed non-monotonic variations with magnetization (and, hence, with temperature) could be detected for the second anisotropy constant, a phenomenon which has not been reported to date. This is a challenge to the experimental work in the field of anisotropy.

To give a feeling of what new features might be expected to arise in cases when three basis functions are involved, in figure 3(a, b) we give canonic plots of all the three anisotropy constants for some arbitrary values of the intrinsic ratios $r_{21} \equiv r = K_2^0/K_1^0$ and $r_{31} = K_3^0/K_1^0$. These ratios are of paramount importance for the variation of the constants upon changes of magnetization or temperature, respectively. (In fact, it would not be exaggerated to claim that the $p - 1$ dimensionless parameters are *constitutional* parameters for the single-ion anisotropy in a given system. It has been demonstrated [8], for instance, that the values of these parameters determine unambiguously the temperature evolution of anisotropy in any given system and corresponding anisotropy flow diagrams have been given which are valid for the whole Callen and Shtrikman class [9].)

Returning to figure 3(a), one may observe the considerable enhancement of the lower-order anisotropy constants K_1 and K_2 which is due to the inclusion of the third-order basis function $\bar{\kappa}_6$. In the presented case all three intrinsic constants K_p^0 ($p = 1, 2, 3$) are equal (i.e., $r_{21} = r_{31} = 1$). An important practical implication is that, should it be possible to synthesize such materials or detect such a property in already existing ones, the first anisotropy constant would be enhanced and its extremum would be pushed towards higher

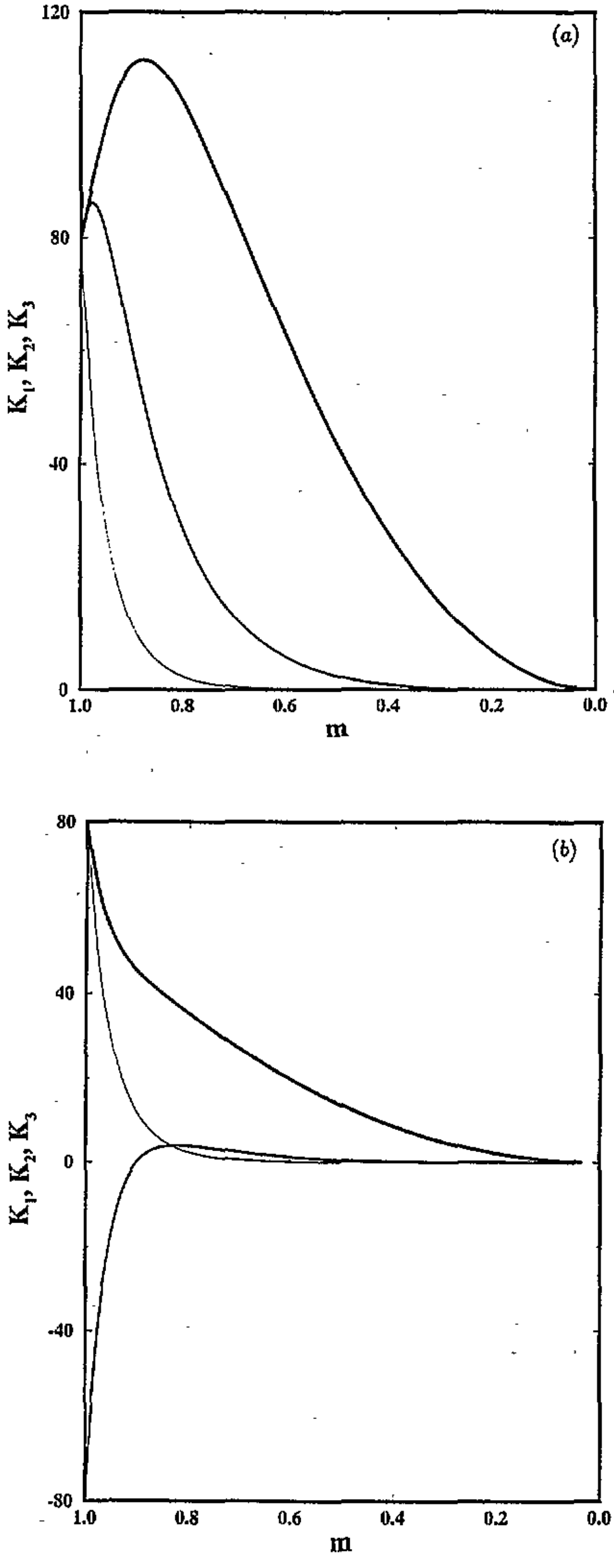

Figure 3. Illustrative canonic dependences for the first three anisotropy constants (in arbitrary units) as functions of magnetization ($J = \frac{9}{2}$). The thickness of lines decreases when going from K_1 to K_3. (a) Enhancement of K_1 and K_2 due to the higher-order coefficients in the three-constant case. Here, signs and magnitudes of the intrinsic anisotropy constants are taken as equal ($K_1^0 = K_2^0 = K_3^0$). (b) Emergence of new features in the canonic dependences in the three-parameter case ($K_1^0 = -K_2^0 = K_3^0$). $K_2(m)$ exhibits a change of sign and a related maximum, while K_1 develops an additional inflexion point when computed as a function of temperature ($K_1(T)$ is not given, since it is not canonic).

temperatures which is an effect of direct practical relevance. In figure 3(b), we have chosen $K_1^0 = K_3^0 = -K_2^0$ (i.e., $r_{21} = -r_{31} = -1$). Interestingly, but not unexpectedly in view of the present analysis, $K_2(m)$ is of the non-monotonic type with a zero point. The canonic curve $K_1(m)$ is monotonic, but it is to be noted that it behaves more peculiarly than in any of the canonic cases in the two-constant anisotropy analysis. We have proceeded to calculate the *temperature* dependence of K_1 within the mean-field approximation with these particular ratios and have found that, indeed, it exhibits a new feature, namely, it changes its curvature three times inside the interval of variation (two inflexion points), whereas only a single inflexion point is possible in the monotonic regime between the borderlines in figure 2 in the two-constant anisotropy case. This observation, though made for a 'non-canonic' dependence (the temperature dependence which is specific for each of the member theories of the Callen and Shtrikman class), implies that the characterization of the generic types has to be refined in the case when more than two anisotropy constants are involved.

The second advance which can be carried through successfully is the adequate treatment of *two-ion* anisotropy. It can be mentioned at this stage that a parametric method is being developed for this case as well; among other things, it makes known analytic results concerning two-ion anisotropy easily applicable.

7. Summary

We have emphasized the role played by the set of fundamental anisotropy coefficients $\bar{\kappa}_n$ in the description of magnetic anisotropy and magnetostriction of single-ion origin. A powerful parametric method allows one to compute the anisotropy coefficients as functions of magnetization. The obtained dependences are valid, and we have correspondingly called them *canonic*, for a whole class of untrivial theories under the assumptions of the Callen and Callen theory of anisotropy and magnetostriction. Expressing the anisotropy *constants* K_i as linear combinations of the basis functions $\bar{\kappa}_n$ via the general relations for the transformation between the two equivalent descriptions of anisotropy energy, one finds the families of canonic curves for the constants as functions of magnetization. The different families are determined by the $p-1$ intrinsic ratios K_i^0/K_1^0 ($i = 2, 3, \ldots, p$), where p is the number of anisotropy constants included in the corresponding anisotropy free energy. It is thus of paramount importance that the intrinsic (constitutional) anisotropies are experimentally measured for each particular case of interest. For the class of theories discussed here and with respect to the variation of anisotropy with magnetization or temperature, their knowledge is as important as the assignment of initial conditions in classical mechanics, for instance. The canonic dependences $K_i(m)$ provide the framework for the determination of the temperature or field dependences by inserting the *experimentally* measured magnetization as a function of temperature or field, respectively, as has indeed been the practice for analysing anisotropy and magnetostriction in rather different experimental situations.

The two-constant case has been completely specified. Three types of canonic dependence for the anisotropy *constants* have been found in both uniaxial and cubic symmetry. Each particular canonic dependence is valid for the whole class of theories for a given value of the intrinsic ratio $r = K_2^0/K_1^0$. A straightforward extension of the method allowed us to predict three types of variation for the magnetostriction of single-ion origin in the two-constant case. By the same token, one finds that in uniaxial symmetry and within the three-constant analysis three generic types of variation are possible for the *second* anisotropy constant K_2 as a function of magnetization or temperature. The way to analyse systematically the three-constant case has been discussed and some further peculiarities (apart from the predictions for the behaviour of K_2) have been illustrated.

We have furthermore suggested that the results of the analysis of the case with two basis functions $\{\bar{\kappa}_n\}$ are applicable as an alternative explanation for the occurrence of near-zero magnetostriction in Co- and Fe-rich amorphous alloys. By now, the compensation has always been viewed as the outcome of counteracting single-ion and two-ion contributions. In contrast, the mechanism suggested in this paper is the competition between single-ion anisotropies of different orders. A necessary step for the analysis from the theoretical side is the development of a parametric method for the computation of the dependence of the *two-ion* anisotropy on magnetization so that a quantitative resolution of the described alternative would be possible. Moreover, such an advance may provide for the simultaneous account of both types of anisotropy, thus transforming the alternative into an interpolation between the one-ion and two-ion mechanisms with different weights. Since physical intuition and some earlier studies indicate that the dependence of the two-ion contributions on magnetization is also monotonic [31], one is once again, as with the case of $p = 3$ for the magnetic anisotropy energy above, led to consider the effect of superposing three different monotonic functions, namely, those for the lowest and next-lowest single-ion anisotropy plus the one describing the two-ion contribution. Unluckily, the latter function could hardly be computed effectively for the whole class, envisaged in this paper, which means that loss of generality seems unavoidable. The testing ground for such developments would be the degree of accuracy of fitting to existing experimental measurements on Co-rich zero-magnetostrictive alloys ([22, 25–30] and references therein). The issue deserves further elucidation.

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References

[1] Callen E and Callen H B 1965 *Phys. Rev.* **139** A455
[2] Birss P R 1965 *Selected Topics in Solid State Physics* vol III, ed E P Wohlfarth (Amsterdam: North-Holland)
[3] Darby M I and Isaac E D 1974 *IEEE Trans. Magn.* **MAG-10** 259
[4] Hutchings M T 1964 *Solid State Physics* vol 16 (New York: Academic) p 227
[5] Elliott R J 1972 *Magnetic Properties of Rare-Earth Metals* ed R J Elliott (New York: Plenum) ch 1, pp 1–16
[6] Millev Y and Fähnle M 1994 *J. Magn. Magn. Mater.* **135** 285
[7] Millev Y and Fähnle M 1995 *Phys. Rev.* B **51** 2937
[8] Millev Y and Fähnle M 1995 *Phys. Rev.* B **52** at press
[9] Callen H and Shtrikman S 1965 *Solid State Commun.* **3** 5
[10] Callen H B 1963 *Phys. Rev.* **130** 890
[11] Tahir-Kheli R A 1975 *Phase Transitions and Critical Phenomena* vol 5B, ed C Domb and M S Green (London: Academic) pp 259–341
[12] Millev Y and Fähnle M 1992 *Phys. Status Solidi* b **171** 499
[13] Millev Y and Fähnle M 1992 *Am. J. Phys.* **60** 947
[14] Jensen J and Mackintosh A R 1991 *Rare Earth Magnetism: Structures and Excitations* (Oxford: Clarendon)
[15] Keffer F 1955 *Phys. Rev.* **100** 1692
[16] Abramowitz M and Stegun I 1965 *Handbook of Mathematical Functions* (New York: Dover) ch 10
[17] Wolf W P 1957 *Phys. Rev.* **108** 1152
[18] Wolf W P 1957 *Air Force Cambridge Research Center Scientific Report* 12
We are grateful to Professor Wolf for providing a copy of the report.
[19] Kazakov A A 1974 *Fiz. Tverd. Tela* **16** 1386 (Engl. Transl. 1974 *Sov. Phys.–Solid State* **16** 893)
[20] Rudowicz C 1985 *J. Phys. C: Solid State Phys.* **18** 1415
[21] Lines M E 1979 *Phys. Rep.* **55** 133

[22] du Tremolet de Lacheisserie E 1993 *Magnetostriction: Theory and Applications of Magnetoelasticity* (Boca Raton, FL: Chemical Rubber Company)
[23] Fulde P 1979 *Handbook on the Physics and Chemistry of Rare Earths* vol 2, ed K A Gschneider Jr and Le R Eyring (Amsterdam: North-Holland) pp 295–386
[24] Landau L D and Lifshitz E M 1959 *Theory of Elasticity (Course of Theoretical Physics 7)* (London: Pergamon)
[25] O'Handley R C 1987 *J. Appl. Phys.* **62** R15
[26] Gonzalez Estevez J and du Tremolet de Lacheisserie E 1990 *J. Phys.: Condens. Matter* **2** 6235
[27] O'Handley R C and Grant N J 1985 *Proc.on Rapidly Quenched Metals* ed S Steeb and H Warlimont (Amsterdam: Elsevier) p 1125
[28] Fähnle M, Furthmüller J, Pawellek R and Beuerle T 1991 *Appl. Phys. Lett.* **59** 2049
[29] Fähnle M, Furthmüller J, Pawellek R and Elsässer C 1988 *Physics of Magnetic Materials* ed W Gorzkowski, K H Lachowicz and H Szymczak (Singapore: World Scientific) p 204
[30] Fähnle M, Furthmüller J, Pawellek R and Herzer G 1989 *Physics of Magnetic Materials* ed W Gorzkowski, K H Lachowicz and H Szymczak (Singapore: World Scientific) p 228
[31] Callen H B and Callen E R 1964 *Phys. Rev.* **136** A1675