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Letter to the Editor

# Anomalous behaviour of the second constant of magnetic single-ion anisotropy as a function of magnetization and temperature

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**Abstract**

An exhaustive study is carried out for the determination of the possible types of variation of the second single-ion anisotropy constant $K_2$. Three types are found and quantified, two of which are anomalous and unknown by now. While they both exhibit a non-monotonic variation with a local extremum between zero and the Curie temperature, one of them is additionally characterized by a zero point and, consequently, by *a change of sign* of $K_2$. The value of a unique parameter (the ratio $K_3^0/K_2^0$) is vital for the classification of the above types, whereby the value of the first anisotropy constant is completely irrelevant. The dependences for the variation of $K_2$ are generated with the help of a new parametric method.

It is well-known [1–4] that magnetic anisotropy can be characterized either by the set of anisotropy constants $K_i$ or by the equivalent set of anisotropy coefficients $\overline{\kappa}_n$. These sets carry the temperature $(T)$ and magnetic-field $(H)$ dependence of the anisotropy energy and it has proved advantageous to express them as functions of the magnetization $m$ which on its part depends on $T$ and $H$. When the anisotropy problem is expressed in terms of the magnetization [4,5], two principal strategies can be pursued. One can either insert the experimentally measured dependences $m(T, H)$ (the semi-phenomenological alternative) or address the problem of computing these dependences within the framework of statistical mechanics (the theoretical alternative). Below we tackle the problem theoretically by using an extension of the approach of Callen and Callen [4,6] to a whole class of untrivial theories [7]. The method allows the computation of the fundamental anisotropy coefficients $\overline{\kappa}_n$ for finite (quantum) values of the total angular momentum $J$ in contrast to the infinite-$J$ (classical) case used previously. The statistic-mechanical calculation to first order in the anisotropy contributions which are treated as perturbations on the dominant exchange interaction is backed up by a powerful parametric approach and has been discussed in sufficient detail in Refs. [8,9].

The relevant information about the experimentally accessible quantities $K_i$ is gained by feeding the information from the calculation of $\overline{\kappa}_n$ into the relations connecting the anisotropy constants $K_i$ with the anisotropy coefficients $\overline{\kappa}_n$ [10,11]. Here we con-

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centrate on the single-ion second anisotropy constant $K_2$ in systems of uniaxial symmetry with negligible in-plane anisotropy. It turns out possible to classify the allowed types of its variation by exploiting the anisotropy coefficients $\bar{\kappa}_n$ as basis functions spanning the physically relevant dependences on $m$, $T$ and $H$. The outcome of this analysis is astonishing, since two of the possible types are quite unusual.

From the relations transforming between the equivalent sets of constants and coefficients in uniaxial systems for the case when the first three anisotropy constants $K_1$, $K_2$ and $K_3$ are relevant [6,10,11], one finds

$$\frac{K_2}{K_2^0} = \left(1 + \frac{18}{11} r'\right) \bar{\kappa}_4 - \frac{18}{11} r' \bar{\kappa}_6, \qquad (1)$$

where $\bar{\kappa}_4$ and $\bar{\kappa}_6$ are to be regarded as normalized basis functions carrying the dependence on the magnetization $m(T, H)$, while $r' \equiv K_3^0/K_2^0$ with $K_i^0$ being the intrinsic (ground-state) anisotropy constants defined as $K_i^0 \equiv K_i(T=0)$. The zero temperature value of $K_1^0$ does not enter the above relation and, consequently, all further conclusions for $K_2$ are independent of it. The basis functions $\bar{\kappa}_n$ can be computed [8] for a whole class of untrivial theories [7]. However, they cannot be cast as explicit functions of $T$ and $H$ or, equivalently, of $m$. A powerful parametric approach which circumvents this major difficulty has been developed [8] and is used here for the analysis of $K_2$. It is based on expressing all relevant quantities as functions of the generalized effective field of Ref. [7] and on using this same field as a sweeping parameter [8,9].

A self-suggesting possibility for the study of $K_2$ would be to generate its dependence on $m$ for exhaustively dense set of values of the constitutional parameter $r'$. However, for the case when only two basis functions $\bar{\kappa}_n$ are involved in a linear superposition of the kind given in Eq. (1), it turns out possible to delineate the allowed types of variation of $K_2$ by simple analytic means. One namely investigates the signs of the first derivative of $K_2$ with respect to $m$ at both ends of variation, i.e. for $m \to 1$ (saturation) and for $m \to 0$ (high-temperature, low-field asymptotics). On using the asymptotics of the coefficients $\bar{\kappa}_n$ in these two limits [9], one then finds the required classification of the types of variation by exhausting the allowed combinations of signs of the first derivative. In this way, we have determined *three possible types* of variation of $K_2(m)$ which are defined according to the value of the unique parameter $r'$ as follows:

I. For $r' > 5/9$, $K_2(m)$ has an extremum which is a minimum or a maximum depending on the sign of $K_2^0$.

II. For $-11/18 < r' < 5/9$, $K_2(m)$ is strictly monotonically decreasing or increasing for $K_2^0 > 0$ or $K_2^0 < 0$, respectively.

III. For $r' < -11/18$, $K_2(m)$ has a zero point at some $m_S$ between zero and saturation; this value can be easily determined with the help of the parametric method. In other words, in this regime the second anisotropy constant changes sign and this is the most striking prediction of the present study.

In fact, both non-monotonic types of variation (type I and type III) are equally surprising and we are not aware of an earlier recognition of such regimes of variation as allowed by single-ion contributions only. Furthermore, the same classification holds for the temperature dependence of $K_2(T)$ in zero external field. The proof is readily furnished by examining the asymptotic expansions of the anisotropy coefficients $\bar{\kappa}_n$ at both ends of the interval of variation and checking that the arising polynomials which depend

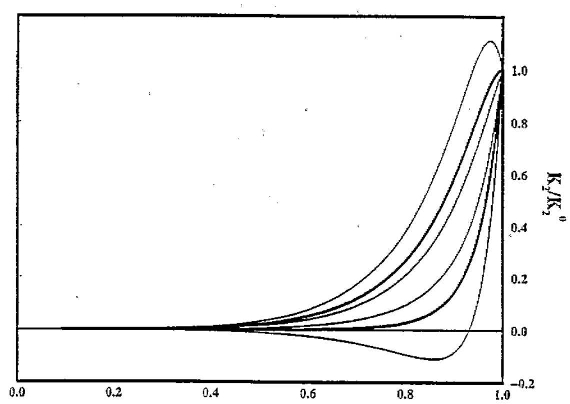


Fig. 1. Types of variation of $K_2(m)$ ($J = 9/2$). From above, the family of curves corresponds to the following values of the constitutional parameter $r' = K_3^0/K_2^0$: 10/9, 5/9, +5/8, −5/8, −11/18 and −11/9. Thick lines denote the borderlines between the three detected regimes of variation.

on the quantum number $J$ do not influence the sign of the derivative of $K_2$ with respect to $T$, i.e. the sign is the same as with the $m$-dependence above. Note also that the classification given above does not depend on $J$, although the basis functions $\overline{\kappa}_n$ do.

The three types of variation of $K_2$ are illustrated in Fig. 1. The dependences presented there are indeed canonic, since they are valid for the whole class of Callen and Shtrikman [7] for a given value of $J$. The thick lines in the figure give the borderlines between the different regimes at $r' = 5/9$ (upper curve) and $r' = -11/18$ (lower curve). The curves correspond to $J = 9/2$. Any other relevant value of $J$ can be treated just as easily. These canonic dependences have been generated with the help of the new parametric method discussed above. From an experimental point of view, the exotic dependences should be readily observable in existing magnets, because the corresponding types I and III are fairly unrestrictive. It is only required that $K_3^0 > (5/9)K_2^0 > 0$ or $K_3^0 < (5/9)K_2^0 < 0$ for the type I to occur and, respectively, $K_3^0 < -(11/18)K_2^0 < 0$ or $K_3^0 > -(11/18)K_2^0 > 0$ for the type III with a change of sign of $K_2$ to occur. In other words, the first type of possible behavior (regime I) would arise by the concerted action of $K_2^0$ and $K_3^0$ having the *same sign* with $|K_3^0|$ sufficiently large, whereas the third type (regime III) sets in because of the competition between these quantities (*opposite signs* with a large enough $|K_3^0|$). There is, however, nothing extreme in the contributions required from the highest-order intrinsic anisotropy constant in either case, especially if one recalls the typical values of the analogous ratio $K_2^0/K_1^0$ in materials where the single-ion anisotropy originates in rare-earth ions sitting on sites of uniaxial symmetry [11,12].

The present study is indicative of the surprises that might be expected in an investigation into the possible types of variation of the *first* anisotropy constant $K_1$ in the three-constant approximation to the free energy. There, however, the situation is much more complicated and the variety of possibilities is founded on a two-fold parametrization with $r = K_2^0/K_1^0$ and $r'$. This case does not allow a conclusive explicit treatment of the type given above and must be tackled separately by searching through the anisotropy space of the system exclusively with the help of the parametric approach and of the related concept of anisotropy flows [9].

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