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December 1974

Prepared for the U. S. Energy Research and Development Administration under Contract W-7405-ENG-48

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UNUSUAL EASY DIRECTIONS OF MAGNETIZATION IN SOME RARE-EARTH IRON CUBIC LAVES COMPOUNDS

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ABSTRACT

Unusual, [uuw] and [uv0]-type easy axes of magnetization have been observed in some cubic rare-earth iron Laves compounds. The phenomenological treatment of the magnetic anisotropy requires the presence of 8th power cosine terms, in order to account for the presence of such directions of magnetization. The conditions imposed on the bulk magnetic anisotropy constants are derived.

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The magnetic anisotropy free energy of cubic materials can be expanded, according to the phenomenological treatment, into a power series of the direction cosines α_i of the axis of magnetization with respect to the crystal axes. Usually only terms up to the 6th power of cosines are retained, thus

$$E_a = K_0 + K_1(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2) + K_2(\alpha_1^2\alpha_2^2\alpha_3^2) \ldots$$

with $\alpha_1^2 + \alpha_2^2 + \alpha_3^2 = 1$, the K_i-s being the bulk magnetic anisotropy constants. It can be easily shown, by differentiation with respect to the angles β and γ ($\beta = \cos^{-1}\alpha_1$, $\gamma = \cos^{-1}\alpha_2$), that the only minima for E_a occur for α_i-s corresponding to the major axes of symmetry of the

cubic system, namely the [001], [011] and [111] directions. Which of these directions becomes an easy axis of magnetization depends on the relative values of K_1 and K_2.

In recent years Mossbauer effect studies have been sucesssfully used, in order to determine the magnetic anisotropy properties of cubic binary rare-earth (R) - iron RFe_2[1] and ternary $R^1_xR^2_{1-x}Fe_2$[2,3] Laves compounds. In several instances we found that the easy axes of magnetization of these compounds were not parallel to any of the major cubic axes symmetry. We observed two main types of such behavior. 1. In ternary mixed compounds of type $R^1_xR^2_{1-x}Fe_2$,[4,5] in the course of spin orientations which took place upon change of either the compositions, or the temperature. 2. In two binary rare-earth iron Laves compounds $CeFe_2$[6] and $SmFe_2$.[7] In most cases the departure of the axis of magnetization for the major axis of symmetry takes place within a relatively wide temperature interval. The purpose of this communication is to show that such cases can be analyzed within the framework of the phenomenological treatment of the magnetic anisotropy energy. The analysis allows to establish the conditions required from the bulk magnetic anisotropy constants, in order that the axis of easy magnetization should deviate from the major axes of symmetry.

Since the retention of the 6th power cosine terms only, yields E_a minima associated with major axes of symmetry, we extend the power expansion to yet another term. Thus we start with

$$E_a = K_0 + K_1(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2)$$

$$+ K_i(\alpha_1^2\alpha_2^2\alpha_3^2) + K_3(\alpha_1^4\alpha_2^4 + \alpha_2^4\alpha_3^4 + \alpha_3^4\alpha_1^4) \qquad (1)$$

The conditions for an extremum in E_a are

$$\frac{\partial E_a}{\partial \beta} = \frac{\partial E_a}{\partial \gamma} = 0 \qquad (2)$$

The extremum is a minimum if at point $(\beta_i, \gamma_i,)$ which satisfies conditions (2)

$$\frac{\partial^2 E_a(\beta_i,\gamma_i)}{\partial \beta^2} > 0 \text{ and } \frac{\partial^2 E_a(\beta_i,\gamma_i)}{\partial \beta^2} > 0$$

and the discriminant is definitely positive i.e.

$$\frac{\partial^2 E_a(\beta_i,\gamma_i)}{\partial \beta^2} \cdot \frac{\partial^2 E_a(\beta_i,\gamma_i)}{\partial \gamma^2} - \left[\frac{\partial^2 E_a(\beta_i,\gamma_i)}{\partial \beta \partial \gamma}\right]^2 > 0 \qquad (3)$$

Applying (2) and (3) to the expression for E_a appearing in (1) we obtain the necessary conditions imposed on the K_i-s in order to obtain minima of E_a. The results indicate that such minima exist for directions of the easy axis of magnetization parallel to the major axes of symmetry and, in addition, for several directions of type [uuw] which

correspond to angles $\beta = \gamma$, and of type [uv0] contained in the (001) plane. The additional directions exist only for $K_3 > 0$. For the sake of conciseness, it is helpful to express K_1 and K_2 in units of K_3, appropriately we define $K_1' = K_1/K_3$ and $K_2' = K_2/K_3$. A straightforward calculation (see for details the Appendix) yields the conditions imposed on the K_i'-s for presence of axes of magnetization other than the major axes of symmetry. The condition for [uuw]-type directions are:

1. $-2 < K_2' < 4$

2. $\dfrac{-(K_2' + 2)^2}{24} < K_1' < \dfrac{K_2' - 1}{2}$

The conditions for a [uv0]-type direction are:

1. $0 > K_1' > -\dfrac{1}{2}$

2. $2 < K_2'$

Figure 1 is a graphical representation in the K_1', K_2' plane of the regions with the different possible axes of magnetization. Within the approximately triangular region ABC, the axis of magnetization is of type [uuw]. Within this region the angle $\theta = \cos^{-1}\alpha_3$, defined as the angle between the direction of magnetization and the [001] axis, lies between 0 and 54.4°. Lines of constant θ have been plotted in the ABC region. The change of θ is continuous only across the AB boundary but θ jumps discontinuously, when crossing the AC or BC boundaries. The cross-hatched region in the neighborhood of A corresponds to a region of local minima of E_a for [uuw] types of magnetization. Such directions will therefore not be stable. Region CED is similar to ABC, in that

the direction of easy magnetization is of type [uuw], the angle θ within this region varies between 54.4 and 90°. Between point E and G there is again a very narrow band corresponding to non-stable (local minima of E_a) axes of type [uuw]. Region DBML is part of the area in which the direction of magnetization is of type [$\underline{uv}$0], i.e, $\theta = 90°$ and $\Phi = \tan^{-1}(v/u)$. Lines of constant Φ have also been plotted in this region, which continues indefinitely towards the right, bounded by the straight lines $K_1' = 0$ and $K_1' = -1/2$.

The variation of the angle θ as function of temperature, deduced from Mossbauer effect measurements in $CeFe_2$ and $SmFe_2$, is shown in Fig. 2. In $SmFe_2$ the direction of magnetization rotates continuously from the [110] axis at 140 K towards the [111] axis at 240 K. In $CeFe_2$ the axis of magnetization is parallel to the [001] direction up to 150 K, above this temperature it changes to type-[$\underline{uuw}$] with $\theta \simeq 20°$. Just below the Curie temperature at 230 K, this angle increases to 30°. In some ternary compounds, such as $Ho_{0.5}Er_{0.5}Fe_2$,[5] the behavior is more complex. With increasing temperature the direction of magnetization goes through the sequence [$\underline{uuw}$] → [110] → [$\underline{uv}$0] → [100].

The phenomenological treatment developed above accounts for all types of behavior. In the case of $CeFe_2$, the values of K_1, K_2 and K_3 vary in such a way with increasing temperature that their projection in K_1', K_2' plane follows the heavy arrow (a) in Fig. 1. For $SmFe_2$ the same projection is represented by arrow (b) which crosses region CED going from region [110] towards region [111]. This occurs during the temperature increase from 140 to 240 K. In $Ho_{0.5}Er_{0.5}Fe_2$ the projection follows reverse arrow (b) at low temperatures and then after the general direction of arrow (c).

Examination of Fig. 1 also indicates that a spin reorientation involving the [111] direction, namely of type [111]⇄[100] or of type [111]⇄[110], will not necessarily take place through a transition region, if K_1 and K_2 are sufficiently large, relative to K_3. On the other hand for a [100]⇄[110] spin reorientation, there will always be a transition region, with axes of magnetization of type [uv0], even for very small values of the bulk magnetic anisotropy constant K_3.

Recently a rhombohedral distortion has been reported for $TbFe_2$.[8] This distortion is probably associated with extremely strong magnetoelastic interaction present in $TbFe_2$ and also in $SmFe_2$.[7] In principle therefore, a distortion from cubic symmetry, if present in $SmFe_2$, would detract from the validity of the use of Eq. 1 in a non-cubic material. This however, would not be the case for $CeFe_2$, which is a ferromagnet with its magnetic anisotropy exclusively due to the iron sublattice. The tetravalent Ce^{4+} ion, has no 4f electrons which may give rise to magnetoelastic interactions leading to a distortion from cubic symmetry. No detectable distortions have been observed in either $HoFe_2$ or $ErFe_2$[8] - it can be assumed that the cubic symmetry is retained in the $(HoEr)Fe_2$ compounds.

The single-ion model, which accounts successfully for the main features of the magnetic anisotropy properties of the rare-earth iron Laves compounds,[3] is unable to explain the presence of [uuw] type axes of easy magnetization. The localized 4f electrons of the rare-earth ions do not yield 8th power cosine terms in the magnetic anisotropy energy expansion. The non-negligible presence of K_3 terms in the power expansion of transition metals has previously been observed in

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the course of careful torque measurements on Ni metal.[10] In the present case they might tentatively be attributed to the intrinsic magnetic anisotropy of the iron sublattice.

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APPENDIX

Starting with Eq. 1, substituting $\alpha_3^2 = 1 - \alpha_1^2 - \alpha_2^2$, changing the notation to $\alpha_1 = \cos\beta$ and $\alpha_2 = \cos\gamma$ and applying conditions (2) for the extremum we obtain

$$\frac{\partial E_a}{\partial \beta} = 2 \sin\beta \cos\beta \, (2\cos^2\beta - 1 + \cos^2\gamma)[K_1 + K_2 \cos^2\gamma - 2K_3 (\cos^4\beta - \cos^2\beta + \cos^2\beta\cos^2\gamma + \cos^4\gamma)] = 0$$

$$\frac{\partial E_a}{\partial \gamma} = 2 \sin\gamma \cos\gamma \, (2\cos^2\gamma - 1 + \cos^2\beta)[K_1 + K_2 \cos^2\beta - 2K_3 (\cos^4\gamma - \cos^2\gamma + \cos^2\gamma\cos^2\beta + \cos^4\beta)] = 0$$

Each derivative is a product of 4 factors. These derivatives will simultaneously satisfy conditions (2) whenever one of the 4 factors (not necessarily the same in the two expressions) will vanish. We distinguish several cases.

1. $\sin\beta = \sin\gamma = 0$

This case corresponds to the <100> axes of magnetization.

2. $\cos\beta = 0$ and $2\cos^2\gamma - 1 + \cos^2\beta = 0$

   or

   $\cos\gamma = 0$ and $2\cos^2\beta - 1 + \cos^2\gamma = 0$

This corresponds to the <110> axes of magnetization.

3. $2\cos^2\gamma - 1 + \cos^2\beta = 0$

   and

   $2\cos^2\beta - 1 + \cos^2\gamma = 0$

Corresponding to the <111> axes of magnetization.

Substituting the values $\cos\beta$ and $\cos\gamma$ in each case in the quadratic form (Eq. 3) yields the limiting values of K_1' and K_2' (assuming $K_3 > 0$) for which the above mentioned major axes of symmetry become easy axes magnetization.

4. The non major axes of easy magnetization are obtained by the vanishing of the 2nd and 4th factors respectively in the 2 derivatives i.e.,

$$\cos\beta = 0$$

$$K_1 + K_2 \cos^2\beta - 2K_3 (\cos^4\gamma - \cos^2\gamma + \cos^2\beta\cos^2\gamma/ + \cos^4\beta) = 0$$

this yields the <uv0> directions, the angle ϕ between the direction of magnetization and the [100] axis being in this case $\sin^2 2\phi = \sin^2 2\beta = -\frac{2K_1}{K_3}$ (again $K_3 > 0$). The magnetic anisotropy free energy $E_{uv0} = \frac{-3}{4}\frac{K_1^2}{K_3}$

5. Finally the vanishing of the 3rd factor of one derivative and the 4th in the second or, the vanishing of both 4th factors i.e.

$$2\cos^2\beta - 1 + \cos^2\gamma = 0$$

and

$$K_1 + K_2 \cos^2\beta - 2K_3 (\cos^4\gamma - \cos^2\gamma + \cos^2\gamma\cos^2\beta + \cos^4\beta) = 0$$

or

$$K_1 + K_2 \cos^2\gamma - 2K_3 (\cos^4\beta - \cos^2\beta + \cos^2\beta\cos^2\gamma + \cos^4\gamma) = 0$$

and

$$K_1 + K_1\cos^2\beta - 2K_3 (\cos^4\gamma - \cos^2\gamma + \cos^2\beta\cos^2\gamma + \cos^4\beta) = 0$$

yields the minima for the <uuw> directions. The angle θ (see Fig. 1) in this case is $\theta = \cos^{-1}(1 - 2\cos^2\beta)$

and
$$\cos\beta = \frac{(K_2 + 2K_3) + \sqrt{(K_2 + 2K_3)^2 + 24 K_1 K_3}}{12K_3}$$

substituting in (3) and taking into account that $1 \geqslant \cos^2\beta \geqslant 0$ we obtain the boundaries of region ABDGECA in Fig. 1. The expression for the energy

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in this case is complicated. Numerical computations show that the shaded area near A and in the narrow strip between G and E the minima for a [uuw] direction are local minima only or in other words, the magnetic ansotropy free energy has lower values in these regions for the major axes of symmetry.

FIGURE CAPTIONS

1. Boundaries of regions with different easy axes of magnetization in the $K_1' = K_1/K_3$ and $K_2' = K_2/K_3$ plane. For details see text.
2. Temperature dependence of the angle of inclination θ of the axis of magnetization with respect to the [001] axis in $CeFe_2$ $SmFe_2$.

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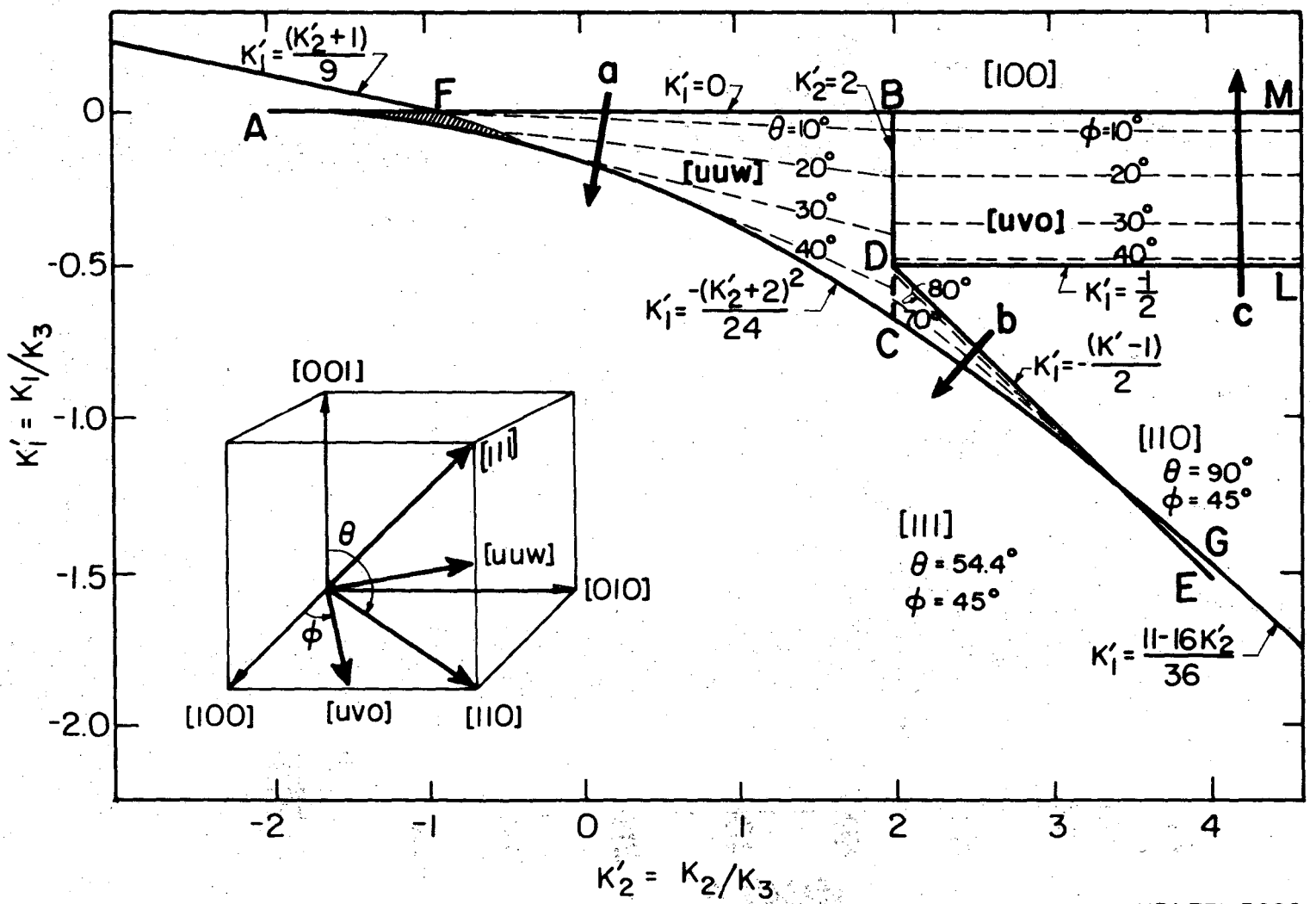

Fig. 1.

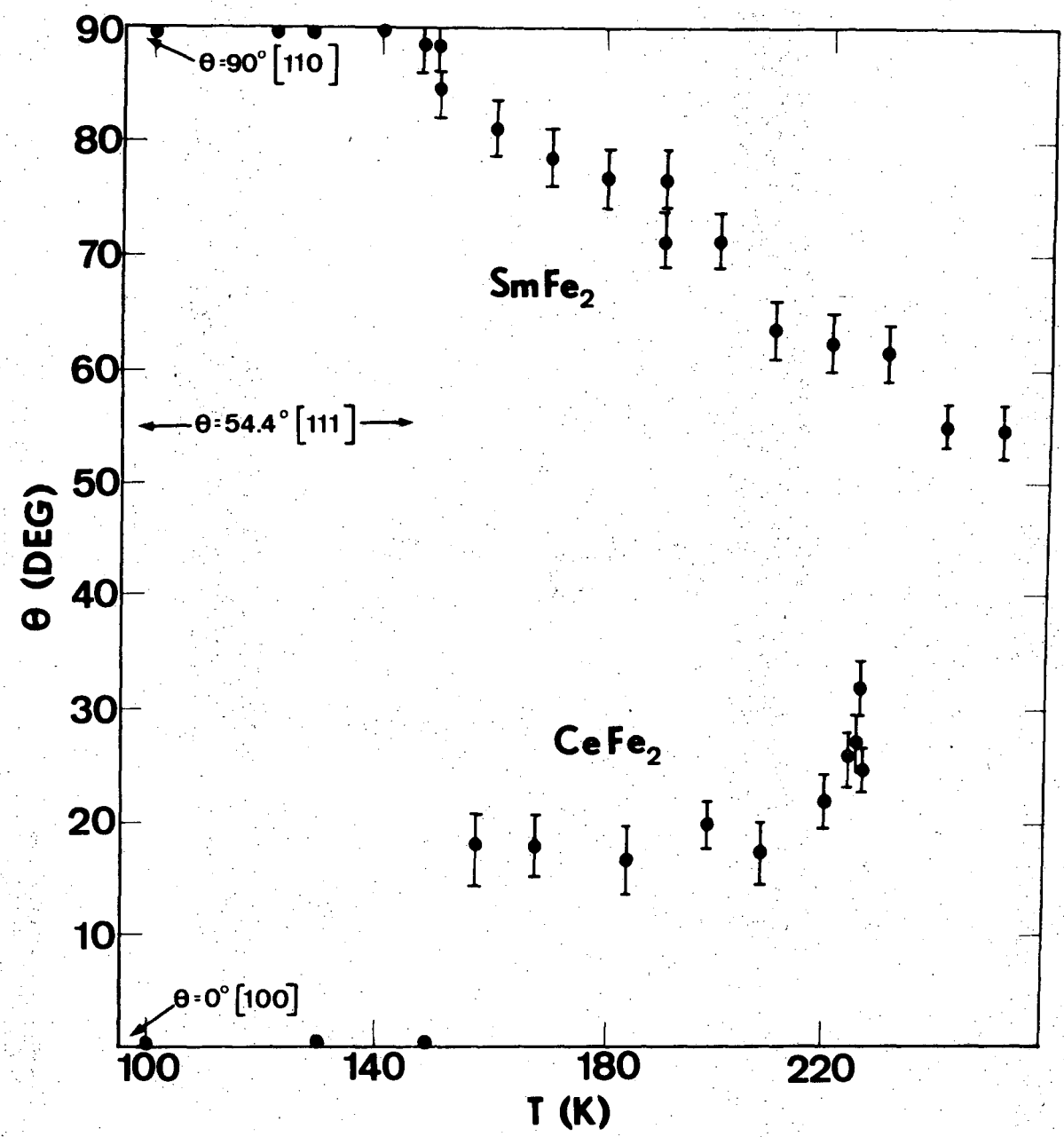

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Fig. 2.

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