Magnetostriction in Dysprosium and Terbium

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The general expressions for single-crystal magnetostriction correct to second order (sixth-rank magnetostrictive tensor) are reduced and evaluated for the particular anisotropies of Dy and Tb. The magnetostrictive constants are determined from x-ray diffraction measurements of the crystal cell distortions. Expressions for the polycrystalline magnetostriction are obtained by suitable averaging and compared with values from the literature.

INTRODUCTION

MAGNETOSTRICTION is the spontaneous distortion or change in dimensions of a material, which is associated with its magnetic behavior.[1] Such dimensional changes may be looked upon as due to the strain dependence of the magnetocrystalline anisotropy energy,[2] or the dimensional and directional dependence of the magnetic exchange energy. Both aspects depend basically on a coupling between the magnetic and elastic energies of the material. The spontaneous strain will depend on both the direction and magnitude of the magnetization. Thus, in ferromagnetic materials with domain structure, two main types of spontaneous magnetostriction may be distinguished. The first and normally more important is the change in dimension upon application of a magnetic field. This anisotropic magnetostriction results from rearrangement of magnetic domains which exhibit an intrinsic distortion, i.e., they no longer possess the symmetry which the crystal would exhibit above the transition temperature of magnetic ordering. The second type is volume magnetostriction, a change in the volume of the crystal cell which depends not on domain structure but on the saturation magnetization. The volume magnetostriction, therefore, exhibits its most pronounced changes in the region just below the magnetic-ordering temperature.

For many materials, in particular, iron, nickel, and cubic alloys of these elements,[1] the magnetostriction dl/l is of the order 10^{-5}. The anisotropic and volume magnetostrictions are then conveniently studied by measurement of dilatation as a function of field and of temperature. For some compounds with antiferromagnetic ordering, e.g., the cubic oxides FeO, CoO, MnO, and NiO, the spontaneous distortion[3,4] is large enough to be observed by x rays as a structure change. This structure change is a lowering of symmetry to tetragonal or rhombohedral below the respective magnetic-ordering temperatures. Although such structure changes result from the same basic interactions, they are not usually considered as magnetostriction since the application of normal fields cause no change in magnetization and, therefore, no dimensional changes.

In the case of ferromagnetic materials, where the spontaneous crystal-cell distortions which give rise to the normal magnetostriction are usually small and not observable by x rays, the material is generally considered to retain its original symmetry. In the case of the cobalt spinel, $CoFe_2O_4$, Guillaud[5] showed that it was possible to observe by x rays the normal magnetostriction in the form of a 0.001-Å dimensional difference in cubically equivalent directions in an oriented polycrystal. With differences as large as this, $\Delta l/l = 1.2 \times 10^{-4}$, it becomes a matter of choice[6] whether one wishes to consider the magnetized crystal in terms of magnetostrictive distortions superimposed upon the original symmetry, or in terms of a new structure of lower symmetry.

Dilatometric measurements[7] of magnetostriction on polycrystalline dysprosium have shown values as large as 2.4×10^{-3}, i.e., nearly an order of magnitude larger than values of magnetostriction for other materials. In the ferromagnetic state of dysprosium, it has been shown[8] that a structure change to orthorhombic takes place. This distortion involves relative changes in linear dimensions as large as 4×10^{-3}. In the demagnetized state the magnetostatic energy causes equal population of moments along each of the six equivalent directions in the basal plane of the original hexagonal structure. The application of a magnetic field favors one of these equivalent directions and results in the observed extraordinarily large dimensional changes. The results of dilatometry and x-ray diffraction are thus different observations of the same phenomenon.

The purpose of this paper is to summarize the expressions for magnetostriction appropriate to the hexagonal structure of dysprosium and terbium, to evaluate the magnetostrictive coefficients from the orthorhombic distortions observed in single-crystal x-ray studies of

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[1] For reviews, see E. W. Lee, Rept. Progr. Phys. **18**, 184 (1955); W. J. Carr, Jr., in *Magnetic Properties of Metals and Alloys* (American Society of Metals, Cleveland, 1959), Chap. 10; R. M. Bozorth, *Ferromagnetism* (D. Van Nostrand, Inc., Princeton, New Jersey, 1951).

[2] C. Kittel, Rev. Mod. Phys. **21**, 541 (1949).

[3] H. P. Rooksby, Acta Cryst. **1**, 226 (1948).

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[5] C. Guillaud, Rev. Mod. Phys. **25**, 64 (1953).

[6] See discussion on cobalt ferrite in R. M. Bozorth, E. F. Tilden, and A. J. Williams, Phys. Rev. **99**, 1788 (1955).

[7] E. W. Lee and L. Alberts, Proc. Phys. Soc. (London) **79**, 977 (1962).

[8] F. J. Darnell and E. P. Moore, J. Appl. Phys. **34**, 1337 (1963).

these metals, and to calculate the magnetostriction expected for polycrystalline samples.

THEORY

The magnetostriction $\lambda=dl/l$ may be written

$$\lambda=S_{ij}\beta_i\beta_j, \tag{1}$$

where S_{ij} are the strains expressed as a second-rank tensor and the β_i are direction cosines of the magnetostrictive strain referred to orthogonal axes. Repeated indexes indicate summation. For a phenomenological approach, the strains may be expanded in a power series in the direction cosines α_i of the magnetization, with suitable use of the requirements of symmetry, which is hexagonal in the present case. Then

$$\lambda=\beta_i\beta_j A_{ijkl}\alpha_k\alpha_l+\cdots$$

and the coefficients A_{ijkl} are determined by comparison with experiment.

An approach which permits a greater physical understanding starts[1] with a crystal energy including terms in magnetocrystalline anisotropy energy, elastic energy, and magnetoelastic energy. In terms of the strains S_{ij}, elastic constants c_{ijkl}, first- , second- , and third-order anisotropy constants K_{mn}, K_{mnop}, and K_{mnopqr}, and first- and second-order magnetostriction terms M'_{ijkl} and N'_{ijklmn}, the energy is

$$\begin{aligned}E=&c_{ijkl}S_{ij}S_{kl}+M'_{ijkl}\alpha_i\alpha_jS_{kl}I_s^2\\&+N'_{ijklmn}\alpha_i\alpha_j\alpha_k\alpha_lS_{mn}I_s^4+K_{mn}\alpha_m\alpha_nI_s^2\\&+K_{mnop}\alpha_m\alpha_n\alpha_o\alpha_pI_s^4+K_{mnopqr}\alpha_m\alpha_n\alpha_o\alpha_p\alpha_q\alpha_rI_s^6.\end{aligned} \tag{2}$$

As shown by Mason,[9] use of only first-order magnetostrictive terms leads to cylindrical symmetry in the hexagonal case, and is, therefore, not adequate for the observations on Dy and Tb. The equilibrium strains S_{ij} are then found[2] from the set of equations

$$\partial E/\partial S_{ij}=0, \tag{3}$$

which give the strains in terms of the constants of the material. Although this approach gives more insight into the significance of the coefficients in the final expression for magnetostriction, in cases where higher order terms must be retained it leads to considerable complexity. To avoid this complexity, Mason[9] uses, instead of E, the thermodynamic enthalpy, $H=E-T_{ij}S_{ij}$, which is a function of the stresses T_{ij} rather than of the strains. The enthalpy function is

$$\begin{aligned}H=&-s_{ijkl}T_{ij}T_{kl}-M_{ijkl}\alpha_i\alpha_jT_{kl}I_s^2\\&-N_{ijklmn}\alpha_i\alpha_j\alpha_k\alpha_lT_{mn}I_s^4+K_{mn}\alpha_m\alpha_nI_s^2\\&+K_{mnop}\alpha_m\alpha_n\alpha_o\alpha_pI_s^4+K_{mnopqr}\alpha_m\alpha_n\alpha_o\alpha_p\alpha_q\alpha_rI_s^6,\end{aligned} \tag{4}$$

where s_{ijkl} are elastic compliances. The matrices M, N are now an inverted form of those designated by primes in (2), and their components are related to the crystal properties in a different way, the exact nature of which will not concern us in this paper. The strain components are now given by

$$S_{ij}=-\partial H/\partial T_{ij}. \tag{5}$$

The magnetostriction is found by substitution of (5) in (1) with the condition $T_{ij}=0$. Thus, the expression for magnetostriction will contain only expressions deriving from energy terms linear in the stresses.

Following Mason, we arrive at the complete expression of magnetostriction for hexagonal anisotropy involving first- and second-order magnetostrictive terms. The orthogonal axes *1*, *2*, and *3* are taken to correspond to hexagonal $[2\bar{1}\bar{1}0]$, $[01\bar{1}0]$, and $[0001]$, respectively.

$$\begin{aligned}\lambda'=&A[2\alpha_1\alpha_2\beta_1+(\alpha_1^2-\alpha_2^2)\beta_2]^2\\&+B\alpha_3^2[(\alpha_1\beta_1+\alpha_2\beta_2)^2-(\alpha_1\beta_2-\alpha_2\beta_1)^2]\\&+C[(\alpha_1\beta_1+\alpha_2\beta_2)^2-(\alpha_1\beta_2-\alpha_2\beta_1)^2]+D(1-\alpha_3^2)\\&\times(1-\beta_3^2)+E\alpha_3^2\beta_3^2(1-\alpha_3^2)+F\alpha_3^2(1-\alpha_3^2)\\&+G\beta_3^2(1-\alpha_3^2)+H\alpha_3\beta_3(\alpha_1\beta_1+\alpha_2\beta_2)\\&+I\alpha_3^3\beta_3(\alpha_1\beta_1+\alpha_2\beta_2)+J\alpha_3^2(1-\beta_3^2)+K\alpha_3^2\beta_3^2.\end{aligned} \tag{6}$$

The following points should be noted: (1) Since the demagnetized state assumed by Mason, with I_s parallel to the *3* axis, is not valid for Dy or Tb, we have replaced terms subtracted by Mason: $M_{31}I_s^2(\beta_1^2+\beta_2^2)+M_{33}I_s^2\beta_3^2+N_{331}I_s^4(1-\beta_3^2)+N_{333}I_s^4\beta_3^2$. This removal of two constraints leads to the addition of the J and K terms, and some changes in the expressions for D and G from those of Mason. (b) The prime on λ' signifies that no demagnetized or ground state has been subtracted. (c) The magnetostriction will be written as $\lambda'(\alpha_1\alpha_2\alpha_3\beta_1\beta_2\beta_3)$ in much of the following in order to make evident the particular restrictive assumptions for α_i and β_i which are possible for Dy and Tb.

The coefficients A, B, etc., involve constants of the material under investigation, the elements M_{ijkl}, etc., corresponding to expression of the magnetostrictive energy in terms of the stresses. Since the subscripts of M_{ijkl} occur in pairs of interchangeable indexes, each pair is replaced by a single index according to the convention

$$1=11;\quad 2=22;\quad 3=33;\quad 4=23;\quad 5=13;\quad 6=12.$$

$$\begin{aligned}A=&(N_{222}-N_{111})I_s^4,\\B=&[3(N_{131}-N_{132}+N_{121})+N_{111}-2N_{222}]I_s^4,\\C=&2(N_{222}-N_{111}-3N_{121})I_s^4+\tfrac{1}{2}(M_{11}-M_{12})I_s^2,\\D=&(2N_{111}-2N_{222}+3N_{121})I_s^4+\tfrac{1}{2}(M_{11}+M_{12})I_s^2,\\E=&[2(N_{111}-N_{222})+3(N_{121}-N_{131}-N_{132}-N_{123})\\&+6N_{133}+N_{331}-N_{333}]I_s^4,\\F=&[2(N_{222}-N_{111})-N_{331}\\&+3(N_{131}+N_{132}-N_{121})]I_s^4,\\G=&3N_{123}I_s^4+M_{13}I_s^2,\\H=&4N_{155}I_s^4+2M_{44}I_s^2,\\I=&4(N_{344}-N_{155})I_s^4,\\J=&N_{331}I_s^4+M_{31}I_s^2,\\K=&N_{333}I_s^4+M_{33}I_s^2.\end{aligned} \tag{7}$$

[9] W. P. Mason, Phys. Rev. **96**, 302 (1954).

Single-Crystal Case

Magnetization[10] and neutron-diffraction[11] studies of Dy have shown that for fields up to 15 kOe the moments are restrained by anisotropy to remain in the plane normal to the *c* or *3* axis. Pulsed-field studies[12] show that fields larger than 100 kOe are required to align moments along the *3* axis. We therefore make the assumption that $\alpha_3=0$. Equation (6) then reduces considerably to

$$\lambda'(\alpha_1\alpha_2O\beta_1\beta_2\beta_3)=A[2\alpha_1\alpha_2\beta_1+(\alpha_1^2-\alpha_2^2)\beta_2]^2+C[(\alpha_1\beta_1+\alpha_2\beta_2)^2-(\alpha_1\beta_2-\alpha_2\beta_1)^2]+D(1-\beta_3^2)+G\beta_3^2. \quad (8)$$

The terms containing D and G are independent of the direction of magnetization and correspond to fixed changes in dimension. As we will see later, it is possible to relate these two terms to observed changes upon ferromagnetic ordering. For application to field-dependent magnetostriction, we should retain only the terms in A and C.

In Dy the easy directions of magnetization[10] are the *1* or $\langle 2\bar{1}\bar{1}0\rangle$. In the demagnetized state, magnetostatic energy considerations and x-ray studies[8] show equal distribution of moments among the six equivalent original hexagonal directions. The magnetostriction for the demagnetized state is obtained by summing (8) over six equal orthorhombic volumes in each of which the magnetization lies along the *1* axis, i.e., $\alpha_1=1$. The summation then requires transformation of the resulting functions of β_i to a single orthorhombic system. This leads to

$$\lambda'(\text{demag})=\tfrac{1}{2}A(1-\beta_3^2)+D(1-\beta_3^2)+G\beta_3^2. \quad (9)$$

The field-dependent magnetostriction in the plane normal to *3* is

$$\lambda(\alpha_1\alpha_2O\beta_1\beta_2O)=\lambda'(\alpha_1\alpha_2O\beta_1\beta_2O)-\tfrac{1}{2}A-D. \quad (10)$$

Parallel magnetostrictions along the *1* and *2* axes are then

$$\lambda(100100)=C-\tfrac{1}{2}A, \quad \lambda(010010)=\tfrac{1}{2}A+C. \quad (11)$$

Transverse magnetostrictions for magnetization along *1* and *2* axes are

$$\lambda(100010)=\tfrac{1}{2}A-C, \quad \lambda(010100)=-\tfrac{1}{2}A-C. \quad (12)$$

These magnetostriction values for $\alpha_2=1$ are not strictly correct for Dy. Magnetization measurements[10] at low temperatures for applied fields parallel to $\langle 10\bar{1}0\rangle$ do not give the saturation obtained for fields parallel to $\langle 11\bar{2}0\rangle$. The moments apparently remain along the nearest $\langle 11\bar{2}0\rangle$ for fields at least up to 8000 Oe. Actually forcing the moment to lie in $\langle 10\bar{1}0\rangle$ directions may lead to a distortion qualitatively different from the orthorhombic cell observed at low fields. We will neglect these corrections since we have, at present, no way of evaluating them.

Polycrystalline Case

We now wish to calculate the longitudinal and transverse magnetostrictions for a polycrystalline sample. If the magnetic field is applied in an arbitrary direction β_i, we assume that the magnetization will remain in the *1*, *2* plane of each crystallite and will saturate in the direction in which the projection of the magnetic field upon the plane is a maximum. The expression for the observable longitudinal magnetostriction, λ_{11}, is found from (8) minus (9) by expression of α_i and β_i in spherical coordinates: $\alpha_1=\cos\phi$, $\alpha_2=\sin\phi$, $\alpha_3=0$, $\beta_1=\sin\theta\cos\phi$, $\beta_2=\sin\theta\sin\phi$, $\beta_3=\cos\theta$.

$$\lambda_{11}=A\sin^2\theta(9\cos^4\phi\sin^2\phi+\sin^6\phi-6\cos^2\phi\sin^4\phi-\tfrac{1}{2})+C\sin^2\theta. \quad (13)$$

Equation (13) must now be averaged over the unit sphere to find the longitudinal magnetostriction for a polycrystalline material of randomly oriented crystallites. It is important to integrate only within one quadrant in order to avoid losing odd functions. The result of averaging is

$$\bar{\lambda}_{11}=\tfrac{2}{3}C. \quad (14)$$

In order to calculate the transverse magnetostriction $\lambda_\perp$, we calculate first the volume magnetostriction ω and then use the relation[1] $\omega=\lambda_{11}+2\lambda_\perp$. We wish to express the volume magnetostriction of a given crystallite in terms of strains along an orthogonal set of axes oriented arbitrarily with respect to the crystallite. We can then average over random orientations to obtain an expression for the polycrystalline case. The new axes *1'*, *2'*, *3'* are related to the crystallite axes *1*, *2*, *3* by the direction cosines:

$$\begin{array}{llll} 1': & \beta_1=\cos\theta\cos\phi, & \beta_2=\cos\theta\sin\phi, & \beta_3=-\sin\theta, \\ 2': & \beta_1=-\sin\phi, & \beta_2=\cos\phi, & \beta_3=0, \\ 3': & \beta_1=\sin\theta\cos\phi, & \beta_2=\sin\theta\sin\phi, & \beta_3=\cos\theta, \\ & \alpha_1=\cos\phi, & \alpha_2=\sin\phi. & \end{array} \quad (15)$$

1' is chosen to lie in the *3*, *3'* plane for simplicity, since we are interested in resolution of the magnetostriction along any set of orthogonal axes. The volume magnetostriction is obtained by summing the expressions for λ given by (8)–(9) evaluated along each of the directions *1'*, *2'*, and *3'*. The terms for *3'* are just λ_{11} calculated above, Eq. (13).

$$\omega=A(9\cos^4\phi\sin^2\phi\cos^2\theta+\sin^6\phi\cos^2\theta-6\cos^2\phi\sin^4\phi\cos^2\theta-\tfrac{1}{2}\cos^2\theta+9\cos^2\phi\sin^4\phi+\cos^6\phi-6\cos^4\phi\sin^2\phi-\tfrac{1}{2})+C(\cos^2\theta-1)+\lambda_{11}. \quad (16)$$

Integrating over random orientations, we have

$$\bar{\omega}=-\tfrac{1}{3}C+\bar{\lambda}_{11}. \quad (17)$$

[10] D. R. Behrendt, S. Legvold, and F. H. Spedding, Phys. Rev. **109**, 1544 (1958).
[11] M. K. Wilkinson, W. C. Koehler, E. O. Wollan, and J. W. Cable, J. Appl. Phys. **32**, 48S (1961).
[12] R. B. Flippen, J. Appl. Phys. **7**, 2026 (1963).

Since $\bar{\omega}=\bar{\lambda}_{11}+2\bar{\lambda}_{\perp}$, (17) yields

$$\lambda_{\perp}=-\tfrac{1}{6}C. \tag{18}$$

Comparison of the single-crystal magnetostrictions, (11) and (12), with the polycrystalline, (14) and (18), shows that preferred orientation in the latter case can have a very large effect.

In Tb the easy directions of magnetization are the b or $\langle 10\bar{1}0\rangle$, as we shall see below. Analysis for the demagnetized state is similar to that preceding (9) except that $\alpha_2=1$ rather than $\alpha_1=1$. Summation over the resulting six equivalent cells gives the result (9) in this case also. The single-crystal and polycrystalline expressions then have the form determined above, although the constants will have different signs.

Dysprosium

In the ferromagnetic state of Dy, x-ray diffraction[8] shows the crystal cell to be orthorhombic. The observed changes in dimensions from the hexagonal or ortho-hexagonal cell to the orthorhombic cell are (at 86°K)

$$\Delta 1/1=0.0025,\quad \Delta 2/2=-0.0042,\quad \Delta 3/3=0.0018. \tag{19}$$

In the present notation these correspond, respectively, to $\lambda'(100100)=C+D$, $\lambda'(100010)=A-C+D$, and $\lambda'(\alpha_1\alpha_2 0001)=G$. We evaluate D, an isotropic change in dimension of the basal plane, from the observed change in area at the 86°K transition

$$\frac{\Delta(1\times 2)}{1\times 2}=-0.0016=\frac{\pi(r+\Delta r)^2-\pi r^2}{\pi r^2}=\frac{2\Delta r}{r}=2D,$$

or $D=-0.0008$. Then $C=0.0033$, $A=0.0001$, and $G=0.0018$. The observable single-crystal magnetostrictions just below 86° will be

$$\lambda(100100)=3.3\times 10^{-3},\quad \lambda(010010)=3.3\times 10^{-3},$$
$$\lambda(100010)=-3.3\times 10^{-3},\quad \lambda(010100)=-3.3\times 10^{-3}. \tag{20}$$

Note that these will change at lower temperatures as described below. The magnetostriction $\lambda(\alpha_1\alpha_2 0001)=G=1.8\times 10^{-3}$ at 86° is a field-independent expansion in the *3* direction which takes place upon transition to the ferromagnetic state through variation in either temperature or field. The volume change at the transition, $\Delta(1\times 2\times 3)/1\times 2\times 3$, is less than 0.0001. This volume change, the difference between the ferromagnetic state and the *antiferromagnetic* state, is not the ordinarily described volume magnetostriction, which is the difference between the ferromagnetic and *paramagnetic* states.

X-ray measurements show that the orthorhombic distortion increases upon further cooling in the ferromagnetic state. With D calculated from the change in area *1×2*, values of A and C can be calculated from the observed $\Delta 1/1$ and $\Delta 2/2$, and such values are shown as a function of temperature in Fig. 1. The determination

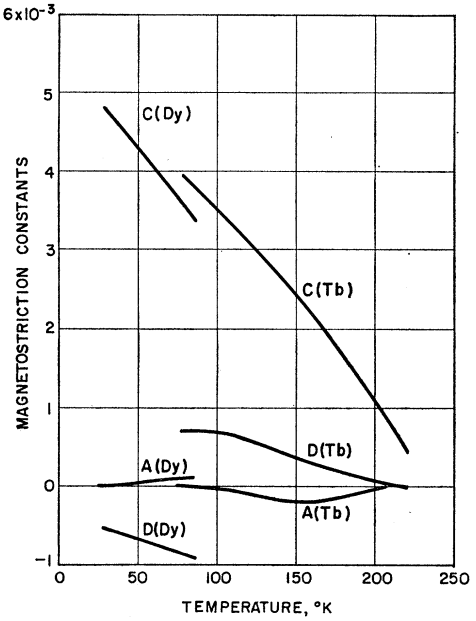

FIG. 1. Magnetostriction constants A, C, and D of Eq. (8) evaluated from x-ray data, for Tb and Dy.

of $\Delta 1$ and $\Delta 2$ involves extrapolation of the antiferromagnetic state from above 86°. This causes the values of A, C, and D to be less accurate at the lower temperatures. The observed *saturation* magnetostriction should increase as the temperature is lowered, with a value of $\lambda(100100)$ at 0°K estimated as 5.5×10^{-3}. At the same time, the demagnetizing and anisotropy fields are increasing, so that magnetostriction in a constant field may actually decrease; such a decrease is reported by Belov *et al.*[13]

Magnetostriction measurements on polycrystalline Dy have been reported by Belov *et al.*[13] and by Lee and Alberts.[7] Both groups found that magnetostriction was still increasing at their highest fields. At 20°K and $H=13\,300$ Oe, Lee and Alberts observed $\lambda_{11}=2.8\times 10^{-3}$ and $\lambda_{\perp}=-1.5\times 10^{-3}$. Substituting from Fig. 1 the value $C=5.0\times 10^{-3}$ into (14) and (18), we obtain $\lambda_{11}=3.3\times 10^{-3}$ and $\lambda_{\perp}=-0.8\times 10^{-3}$. The agreement is not very satisfactory; since the calculated values assume no moment out of the basal plane and use zero-field x-ray data for which the moment lies only in $\langle 2\bar{1}\bar{1}0\rangle$ directions, better agreement should perhaps not be expected.

At 83°K and 15 000 Oe, Belov *et al.* observed $\lambda_{11}=1.1\times 10^{-3}$ and $\lambda_{\perp}=-0.3\times 10^{-3}$. Substituting the value $C=3.4\times 10^{-3}$ into (14) and (18), we obtain $\lambda_{11}=2.3\times 10^{-3}$ and $\lambda_{\perp}=-0.6\times 10^{-3}$. The agreement is again only qualitative. Some degree of preferred orientation, or effects due to impurities which decrease the effective saturation, may be responsible for the low measured values.

[13] K. P. Belov, R. Z. Levitin, S. A. Nikitin, and A. V. Ped'ko, Zh. Eksperim. i Teor. Fiz. **40**, 1562 (1961) [translation: Soviet Phys.—JETP **13**, 1096 (1961)].

Magnetostrictions observed in the antiferromagnetic state are comparable to those observed in the ferromagnetic state. This is understood when it is realized that small fields less than the demagnetizing fields are sufficient to cause transition to the ferromagnetic state. For instance, at 120°K the field required to cause transition to the ferromagnetic state is only ~4000 Oe while the demagnetizing field for a sphere with $M=2310$ emu/cm^3 would be ~10 000 Oe. Measurement of the field-induced magnetostriction in the antiferromagnetic state above 86°K will show first the anisotropic distortion corresponding to terms in D and G of (8), followed by ferromagnetic magnetostriction as analyzed above. The observed magnetostriction will thus be comparable in magnitude to that in the ferromagnetic state below 86°K, although, of course, it will approach zero at the Néel temperature.

Legvold, Alstad, and Rhyne[14] have recently reported single-crystal magnetostriction measurements for Dy. In the antiferromagnetic state at 101°K they find saturation for fields greater than 15 000 Oe applied in the basal plane. The zero-field state in this case is not that described by (9), but the antiferromagnetic hexagonal state of zero strain. In the present notation their values are $\lambda'(100100)=2.2\times10^{-3}$ and $\lambda'(010100)=-3.5\times10^{-3}$. Extrapolation of values in Fig. 1 gives $D=-0.9\times10^{-3}$ and $C=2.9\times10^{-3}$. We then estimate $\lambda'(100100)=C+D=2.0\times10^{-3}$ and $\lambda'(010100)=-C+D=-3.8\times10^{-3}$, in good agreement with Legvold *et al.*

The volume magnetostriction ω may be estimated from x-ray data by comparing the crystal cell volume in the *ferromagnetic* state at 80°K, for example, with the volume obtained by extrapolation of cell volume in the *paramagnetic* state above 180°K. This gives a value $\omega=2.8\times10^{-3}$.

Terbium

Neutron diffraction measurements[15] show that the moments lie in the basal plane of Tb as they do in Dy. The magnetostriction expression (8), therefore, applies to Tb. From x-ray diffraction,[16] which shows expansion in the ferromagnetic state along the $\langle10\bar{1}0\rangle$ directions, and from the observed positive longitudinal magnetostriction,[17] we conclude that the easy directions of magnetization in Tb are the $\langle10\bar{1}0\rangle$, in contrast to the $\langle2\bar{1}\bar{1}0\rangle$ of Dy. This is in agreement with single-crystal magnetization data of Hegland, Legvold, and Spedding.[18] Such a result is predicted by Elliott[19] from crystal-field considerations.

This easy direction requires that in calculating the demagnetized state we sum $\lambda_i'(010\beta_1{}^i\beta_2{}^i\beta_3{}^i)$ rather than the $\lambda_i'(100\beta_1{}^i\beta_2{}^i\beta_3{}^i)$ used above. Evaluation of the required sum leads to the same result for the demagnetized state, (9), that was obtained previously. The single-crystal and polycrystalline expressions derived above will then be valid for Tb.

The observed distortions in Tb, $\Delta1/1$ and $\Delta2/2$, the differences between the ferromagnetic orthorhombic crystal lattice and the paramagnetic hexagonal lattice extrapolated from above 230°K, correspond, respectively, to

$$\lambda'(010100)=-C+D \text{ and } \lambda'(010010)=A+C+D. \quad (21)$$

We again calculate D from the change in area of the basal plane, which in this case is positive and has at 80° a value

$$2D=\Delta(1.2)/1.2=1.4\times10^{-3}, \quad \text{or} \quad D=0.7\times10^{-3}.$$

Values of D, C, and A calculated from the x-ray data[16] are shown in Fig. 1. At 80°, for example,

$$C=3.9\times10^{-3}, \quad A=0.0\times10^{-3}. \quad (22)$$

The single-crystal magnetostrictions observable at 80° will be

$$\begin{aligned}&\lambda(100100)=3.9\times10^{-3}, \quad \lambda(010010)=3.9\times10^{-3},\\ &\lambda(100010)=-3.9\times10^{-3}, \ \lambda(010100)=-3.9\times10^{-3}.\end{aligned} \quad (23)$$

Belov *et al.*[17] have reported magnetostriction measurements on Tb from 85° to 200°K. As in the case of Dy they do not obtain saturation up to 15 000 Oe. Their maximum observed values at 85° are $\lambda_{II}=0.8\times10^{-3}$ and $\lambda_\perp=-0.5\times10^{-3}$. Substituting the value of C from (22) into (14) and (18), we obtain $\lambda_{II}=2.6\times10^{-3}$ and $\lambda_\perp=-0.6\times10^{-3}$. The lack of agreement again may arise from preferred orientation in the sample studied, or from breakdown of some assumption in the present calculations such as saturation, or the restriction of moments to the basal plane. Single-crystal measurements should be more definitive in establishing the validity of the present magnetostriction analysis.

The magnetostriction increases as the temperature is lowered, with an estimated maximum value of $\lambda(010010)$ at 0°K of $C-\frac{1}{2}A=5.1\times10^{-3}$. The volume magnetostriction at 80°K, estimated as in the Dy case, is $\omega=4.6\times10^{-3}$.

We observe for both Dy and Tb that $|A|<0.2\times10^{-3}$. While this still represents a large strain or magnetostriction, it is small compared to values of C and D, and within the errors caused by the necessity of extrapolating lattice constants for the hexagonal state from above the magnetic ordering temperatures. If it is concluded that A is actually zero, then we may deduce

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from (7) that $N_{222}=N_{111}$, and the expressions for C and D become simplified. Such conclusions regarding the magnetostriction tensor components should await more accurate measurements by dilatometric methods, and A, C, and D are here considered as strictly phenomenological constants.

SUMMARY AND CONCLUSIONS

The magnetostriction expressions of Mason for hexagonal symmetry, correct to second order, have been applied to dysprosium and terbium. The negative anisotropy of these metals and the requirement of extremely large fields to turn magnetization out of the basal plane allow the general expression to be considerably simplified. The constants of the simplified relation are evaluated from x-ray measurements of the crystal-cell dimensions of dysprosium in its ferromagnetic state below 86°K, and for terbium in its ferromagnetic state below 220°K. The maximum observable single crystal magnetostrictions are estimated to be $\sim 5\times 10^{-3}$ for both dysprosium and terbium. On the basis of the observed orthorhombic cell in the ferromagnetic state and the observed positive magnetostriction, it is concluded that the easy directions in terbium are the $\langle 10\bar{1}0\rangle$. The volume magnetostriction at 80°K is found from the x-ray cell parameters to be 2.8×10^{-3} for dysprosium and 4.6×10^{-3} for terbium.

Polycrystalline magnetostriction expressions are obtained by suitable averaging of the single-crystal expressions over arbitrary direction of measurement. The calculated values are found to be in qualitative agreement with those for Dy observed by Lee and Alberts and those for Dy and Tb observed by Belov *et al.* Both of these literature measurements were made on presumably unoriented polycrystalline material with fields of $\sim 15\,000$ Oe.

The other heavy rare earths also exhibit large crystalline anisotropies and may be expected to show correspondingly large saturation magnetostriction. However, in Ho and Er the ferromagnetic state retains a spiral configuration in projection on the basal plane, and in zero field will not show any distortion from hexagonal. Large magnetostrictions will occur only for applied fields large enough to collapse the spiral and allow orthorhombic distortion in the plane.

Note added in proof. Magnetostriction measurements on single crystals of dysprosium have recently been reported by Legvold *et al.*[20] and by Clark *et al.*[21] Their data are in good agreement with the present measurements as shown in the following table:

Temp. (°K)	Magnetostriction	Legvold *et al.* ($\times 10^{-3}$)	Clark *et al.* ($\times 10^{-3}$)	This work ($\times 10^{-3}$)
22	$\lambda(100100)$	4.6		5.0
	$\lambda(100010)$	−4.7		−5.0
85	$\lambda(100100)$	3.3	3.8	3.3
	$\lambda(100010)$	−3.1		−3.3
	$\lambda(\alpha_1\alpha_2 0001)$[a]	1.8	1.7	1.8

[a] This is the spontaneous expansion of c associated with transition to the ferromagnetic state.

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