Magnetostriction of Erbium Single Crystals*

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The magnetostriction of Er single crystals has been measured from 300 to 10°K in applied fields up to 30 kOe. In contrast to the other rare earths, the magnetostriction of Er is dominated by field-induced modifications of the exchange energy. The exchange magnetostriction which results from the application of a c-axis field has been determined in the quasi-antiphase-domain and modulated-moment temperature regions. Below 20°K, application of a field in the basal plane produced a sharp change in strain at 18 kOe. This is presumed to arise from a breakdown of the conical ferromagnetic structure into a conical fan state. The second-order anisotropic basal-plane magnetostriction constant was measured. Negative values were found above 55°K in accordance with theoretical predictions. Below 18°K anomalously small and positive values were obtained. The temperature dependence of the a-, b-, and c-axis strains in zero field and in a 30-kOe field was evaluated.

I. INTRODUCTION

SPONTANEOUS and field-induced geometrical distortions (magnetostrictions) are observed in magnetic materials as a result of a coupling between the elastic energy and the magnetic anisotropy and exchange energies. In the heavy rare-earth elements, Tb-Er, these magnetoelastic effects are quite large, principally as a consequence of the large twofold and sixfold anisotropy energies.

Previous magnetostriction studies at this laboratory on single crystals of Dy,[1] Ho,[1] and Tb [2] show that orthorhombic distortions of the hexagonal axes as large as nine-tenths of a percent are produced by rotation of the magnetization through 90° in the basal plane. The exchange magnetostriction of the c axis resulting from the field-induced collapse of the helical magnetic state in Dy was found to be as great as 0.25%.

This paper presents the results of a study of the magnetostriction of Er single crystals in the temperature range from 300 to 10°K in applied fields up to 30 kOe. Linear strains were measured using Budd-type S-421 electrical resistance strain gauges. Details of the apparatus, crystal preparation, and experimental procedure have been discussed previously.[2]

II. MAGNETIC STRUCTURE OF Er

The crystal structure of Er is hexagonal close packed. The orthogonal coordinates used to describe the unit cell are the $a\langle 11\bar{2}0\rangle$, the $b\ \langle 10\bar{1}0\rangle$, and the $c\ \langle 0001\rangle$ axes.

In zero applied field, Er exhibits three distinct ordered magnetic phases, as observed by Cable *et al.*[3] in neutron-diffraction studies. As shown in Fig. 1, the c-axis component of the magnetization is observed to be sinusoidally modulated from 85 to 53°K. No ordering is found in the basal plane. Below 53°K the sinusoidal variation changes to a square-wave modulation, and helical ordering is observed in the basal plane (a quasi-antiphase-domain structure). At about 20°K (17.5°K in this study) the anisotropy energy spontaneously effects a transition to conical ferromagnetism in which the easy magnetic directions are the generators of the cones. At 4.2°K the c-axis component of the moment is 7.2 μ_B and the basal-plane component is 4.1 μ_B. This suggests that the moments lie on a cone of half-angle about 30°.

The axial anisotropy energy producing the conical ferromagnetic state is quite large and only small modifications are possible using normal laboratory fields. Magnetic-moment studies by Green *et al.*[4] in fields up to 18 kOe applied along the c axis yielded a value of the c-axis magnetic moment at 4°K of 7.9 μ_B. This was within 0.7 μ_B of the zero-field neutron-diffraction result. Measurements of the magnetostriction at 10°K with a 30-kOe field applied along the c axis gave values of -4.2 and $+14.0$ (in.$\times 10^{-5}$/in.) for the a- and c-axis strains, respectively, relative to the demagnetized state. These strains are comparatively quite small, again indicating little change in the magnetic state.

III. EXCHANGE MAGNETOSTRICTION IN Er

The change from the quasi-antiphase-domain structure to conical ferromagnetism which occurs spontane-

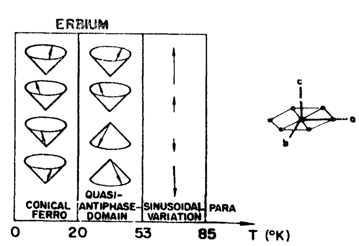

FIG. 1. Magnetic structure of Er in zero applied field observed by neutron diffraction (see Ref. 3).

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[1] S. Legvold, J. Alstad, and J. Rhyne, Phys. Rev. Letters **10**, 509 (1963).
[2] J. J. Rhyne and S. Legvold, Phys. Rev. **138**, A507 (1965).
[3] J. W. Cable, E. O. Wollan, W. C. Koehler, and M. K. Wilkinson, J. Appl. Phys. **32**, 49S (1961).
[4] R. W. Green, S. Legvold, and F. H. Spedding, Phys. Rev. **122**, 827 (1961).

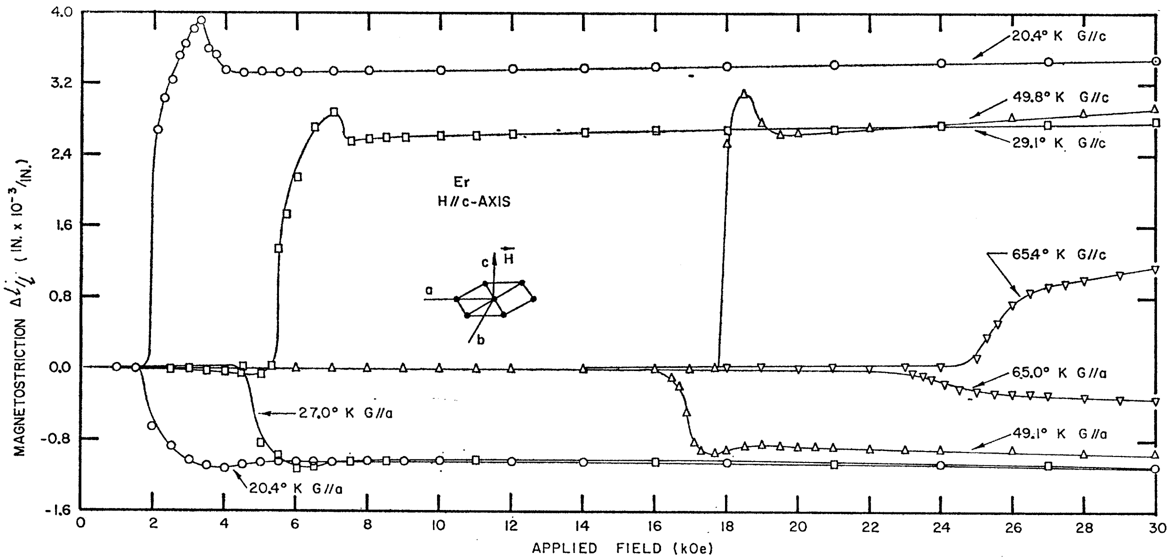

FIG. 2. Magnetostriction of Er as a function of field applied along the c axis. Temperatures below 53°K correspond to the quasi-antiphase-domain structure in zero applied field. The 65°K curves are in the modulated-moment phase. G indicates the strain gauge direction.

ously at 18°K may be induced at temperatures above 20°K by the application of a field along the c axis. The a- and c-axis strains resulting are shown in Fig. 2. Only small strains are observed up to the critical field required to overcome the exchange energy. The critical fields exhibit an almost linear increase with temperature. Complete c-axis ferromagnetic alignment is not possible in the fields used because of the anisotropy. It is noted from the figure that the induced change in the exchange energy results in a large expansion of the c axis and a smaller contraction of the basal plane (the a- and b-axis strains are almost identical on the scale shown in the figure).

Figure 3 shows the c-axis and basal-plane strains at 10°K as a function of field applied along a basal-plane direction. The presence of the large c-axis strain indicates that these magnetostrictions are again dominated by changes in the exchange energy and not by the sixfold basal-plane anisotropy as is the case in other rare-earth ferromagnets. Negligible strain, corresponding to no modification of the magnetic structure, was observed up to 18.5 kOe, at which point a fairly large strain occurred in all three axes. The basal-plane directions were observed to contract accompanied by a large c-axis expansion and a resulting increase in total volume. The critical field of 18.5 kOe was essentially independent of temperature from 15 to 4.2°K and independent of the direction of the applied field in the basal plane. The strain measured perpendicular to the field direction exhibited a larger variation above the "knee" of the curve than that measured parallel to the field direction.

Flippen[5] has measured the magnetic moment of an a-axis Er crystal in static fields up to 56 kOe. He reported a value of 3.8 μ_B along the field direction which is somewhat lower than the 4.1 μ_B reported by Cable *et al.* for the basal-plane component from neutron-diffraction measurement in zero applied field. This fact, together with the observed behavior of the magnetostriction, leads one to the postulate shown in Fig. 4 about the effect of a basal-plane applied field on the magnetic structure of Er below 19°K. It is conjectured that the application of a basal-plane field above about 18 kOe partially overcomes the exchange field and breaks the conical magnetic-moment distribution (a) down into a "fan-like" configuration (b) about the field direction. The relatively larger variation of magnetostrain perpendicular to the field direction suggests that the spread of the fan may be smoothly decreased by increased fields in opposition to the exchange force. At fields higher than present measurements extend, the fan may disappear in favor of ferromagnetic alignment in the plane containing the c axis and the applied field direction (c). Conceivably this could also be accompanied by some modification in the apex angle of the cone in opposition to the large axial anisotropy energy. Eventually, fields large enough to overcome the anisotropy would produce ferromagnetic alignment in the basal plane along the field direction (d). A similar model containing additional intermediate states has been predicted from theoretical calculations by Kitano and Nagamiya.[6]

The magnetostriction resulting from application of a basal-plane field is shown in Fig. 5 in the quasi-

[5] R. B. Flippen, J. Appl. Phys. 35, 1047 (1964).

[6] Y. Kitano and T. Nagamiya, Progr. Theoret. Phys. (Kyoto) 31, 1 (1964).

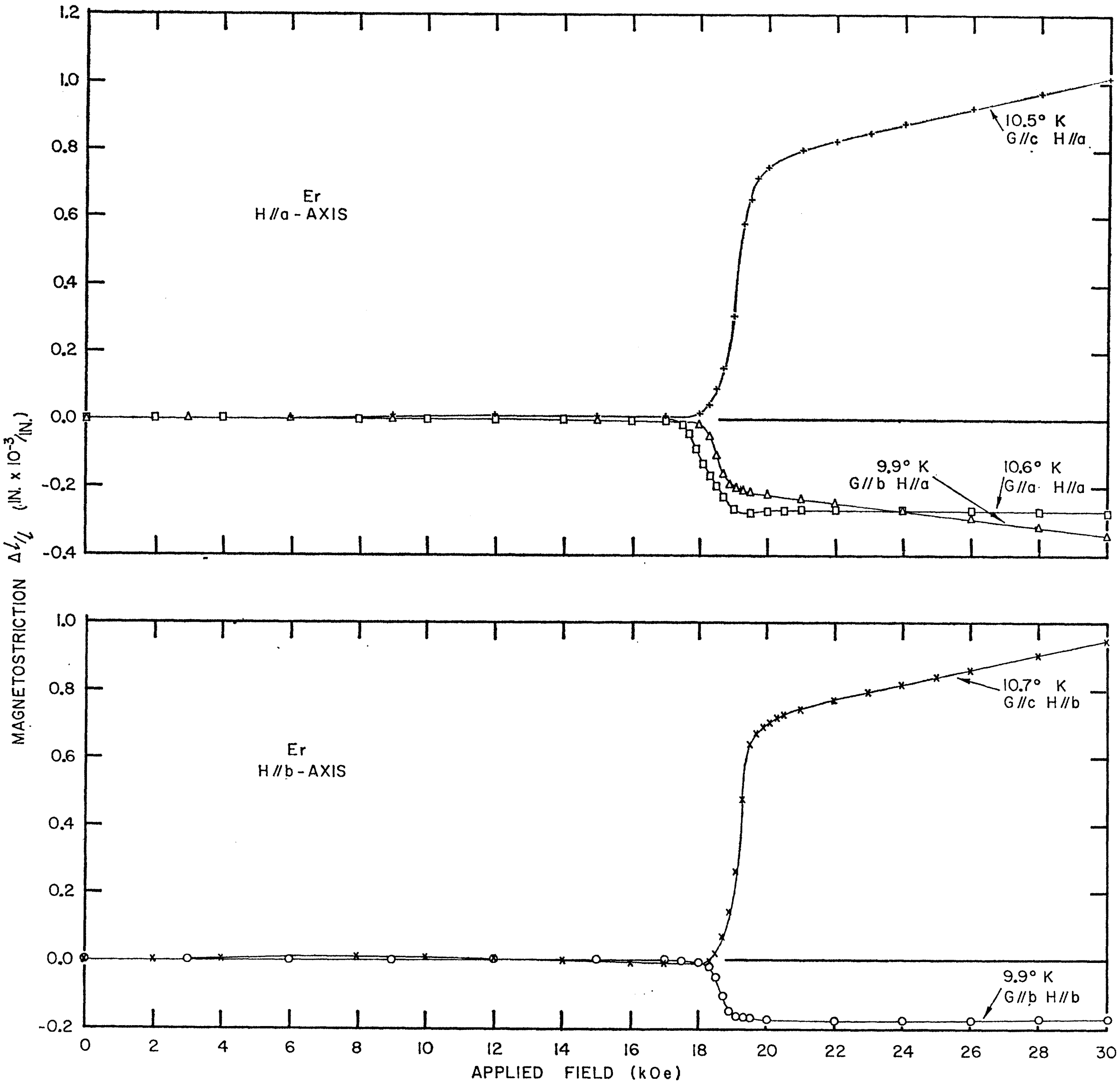

FIG. 3. Magnetostriction of Er as a function of applied field in the conical-ferromagnetic temperature range. The field was applied along the a axis in the upper figure and along the b axis in the lower figure.

antiphase-domain region. The figure indicates considerable modification of the magnetic state is induced by the field. An initial expansion is observed along the field direction accompanied by a transverse contraction. This initial behavior is proportional to H^2. A reversal in slope occurs at higher fields. The parallel magnetostrictions along either the a or b axis are seen to be equivalent after correcting for the difference in measurement temperature.

The results of the magnetostriction measurements in the sinusoidal c-component temperature range are shown in Fig. 6 for the field applied along the a axis. The strain is proportional to the square of the applied field, showing that no change in the state of magnetic order is produced by the application of a field in the basal plane at this temperature.

In the paramagnetic temperature range ($T>85°\mathrm{K}$) the magnetostriction was again proportional to H^2, as shown in Fig. 7 for the field applied along the c axis. Similar H^2 dependence was obtained for the magnetic field along the b axis.

IV. MAGNETOSTRICTION CONSTANTS

Callen and Callen[7] have shown that the magnetostriction of a material may be described by a Hamiltonian containing the elastic energy, one-ion and two-ion

[7] E. R. Callen and H. B. Callen, Phys. Rev. **129**, 578 (1963).

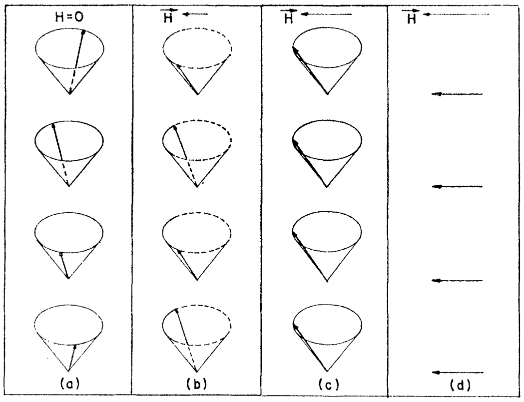

FIG. 4. Possible modifications of the conical ferromagnetic state under the influence of progressively higher fields applied in the basal plane.

magnetoelastic coupling terms which represent, respectively, the strain dependence of the anisotropy energy and the strain dependence of the exchange energy and dipole-dipole energy. This Hamiltonian is then expanded in the symmetry group of the lattice. The free energy is calculated and minimized with respect to the strains to obtain the equilibrium magnetostrains. These strains are transformed back to Cartesian symmetry, and the linear magnetostriction in a direction characterized by direction cosines β is given by $\Delta l/l=\sum_{i,j}S_{ij}\beta_i\beta_j$. The S_{ij} are the equilibrium magnetostrains. The resulting expression can be combined into one containing temperature- and moment-dependent magnetostriction constants λ, multiplied by polynomials of the magnetization direction cosines α_i and the strain direction cosines β_i. Specification of the magnetostriction in Tb [1] and in Dy requires terms up through fourth order in the magnetization direction cosines. Because of the smaller basal-plane anisotropy in Er, the second-order theory corresponding to cylinderical symmetry is sufficient. The second-order magnetostriction expression obtained by the Callens is

$$\begin{aligned}\Delta l/l=&[\lambda_1^{\alpha,0}+\lambda_1^{\alpha,2}(\alpha_z^2-\tfrac{1}{3})](\beta_x^2+\beta_y^2)\\&+[\lambda_2^{\alpha,0}+\lambda_2^{\alpha,1}(\alpha_z^2-\tfrac{1}{3})]\beta_z^2\\&+\tfrac{1}{2}\lambda^{\gamma,2}[(\alpha_x\beta_x+\alpha_y\beta_y)^2-(\alpha_x\beta_y-\alpha_y\beta_x)^2]\\&+2\lambda^{\epsilon,2}(\alpha_x\beta_x+\alpha_y\beta_y)\alpha_z\beta_z.\end{aligned}\quad(1)$$

The x axis corresponds to the a crystal axis, the y axis to the b direction, and the z axis to the c direction. The magnetostriction constants λ are functions of the elastic constants, the magnetoelastic coupling coefficients, and expectation values of the spin operators.[8] The first superscript on the magnetostriction constants λ represents the strain mode as illustrated by Clark *et al.*[8] The constants α denote a symmetry-preserving dilatation of the basal-plane and c-axis directions, the γ constant denotes an orthorhombic distortion of the basal plane, and the ϵ constants represent a shearing of planes perpendicular to the c axis. The second superscript denotes the order l of the associated magnetization direction cosines.

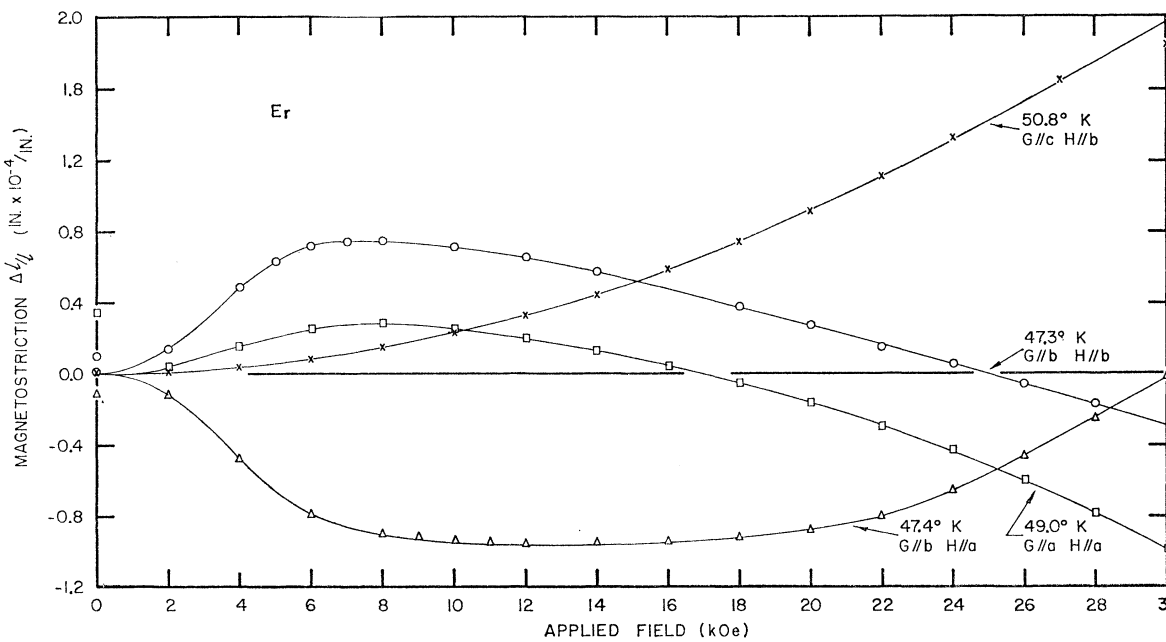

FIG. 5. Magnetostriction of Er as a function of applied field in the quasi-antiphase-domain temperature region. The field was applied in the basal plane. G indicates the strain gauge direction.

[8] A. E. Clark, B. F. DeSavage, and R. M. Bozorth, Phys. Rev. 138, A216 (1965).

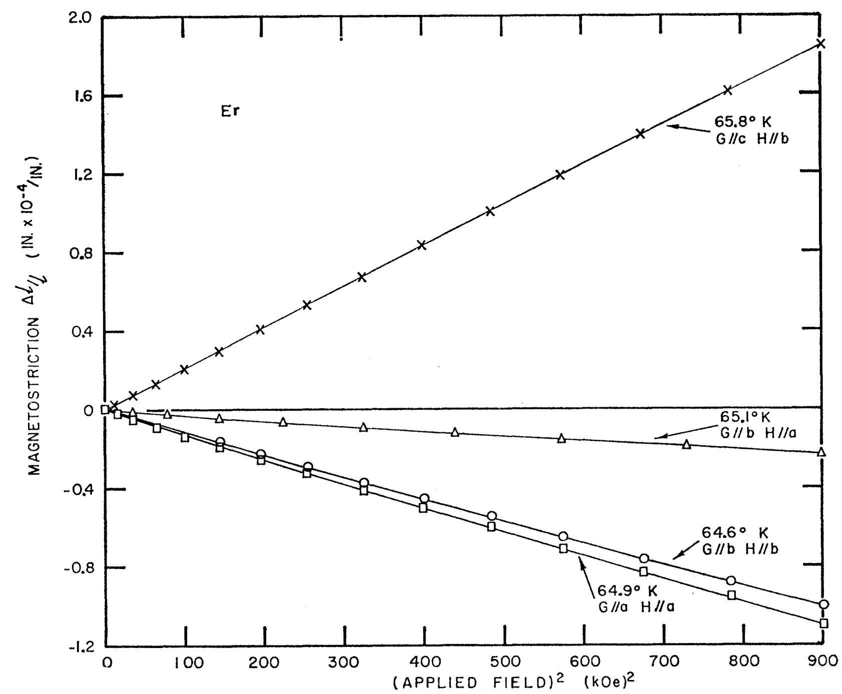

FIG. 6. Magnetostriction of Er as a function of H^2 applied in the basal plane. The curves shown are in the modulated-moment temperature range. G indicates the strain gauge direction.

In Er, because of incomplete knowledge of the magnetic states above 20°K in the presence of an applied field and because of the extreme anisotropy, no attempt was made to evaluate any of the magnetostriction constants except $\lambda^{\gamma,2}$. This constant was obtained from measurements of the a- or b-axis strain as the magnetic field was rotated through 90° in the basal plane. If the magnetization is constrained to a cone of half-apex angle ϕ, the expression obtained for the a- or b-axis strain as a function of applied field angle θ_a' relative to the a axis is

$$(\Delta l/l)\,|_{a,b}{}^{\theta_a'} = \mp\lambda^{\gamma,2}\sin^2\phi\,\sin^2\theta_a'. \tag{2}$$

The prime indicates that the strain of the reference state has been subtracted. Figure 8 shows data for the b-axis strain as a function of applied field angle in the basal plane relative to the a axis. Results are shown in both the paramagnetic ($T>85$°K) and sinusoidal mo-

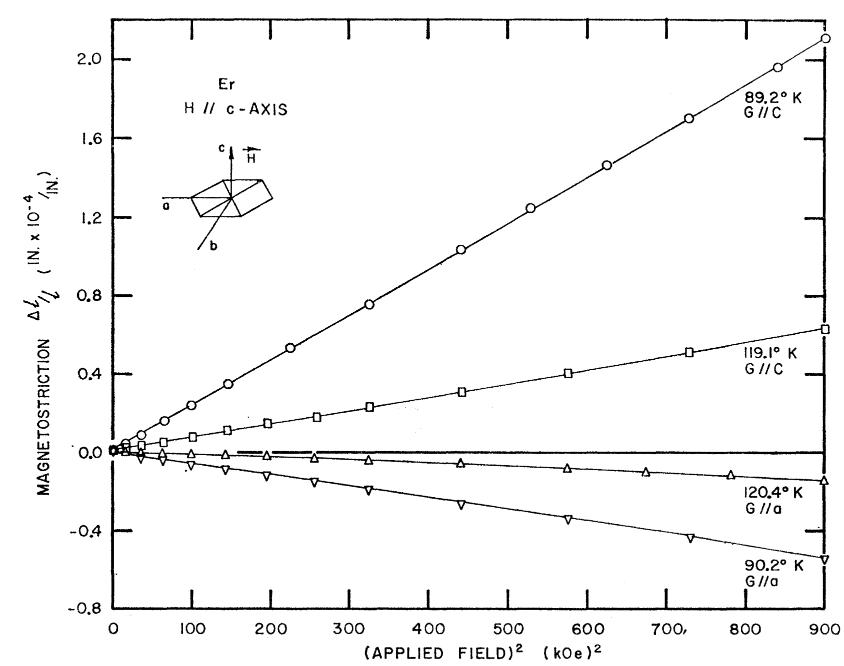

FIG. 7. Magnetostriction of Er in the paramagnetic temperature range as a function of H^2 applied along the c axis. G indicates the strain gauge direction.

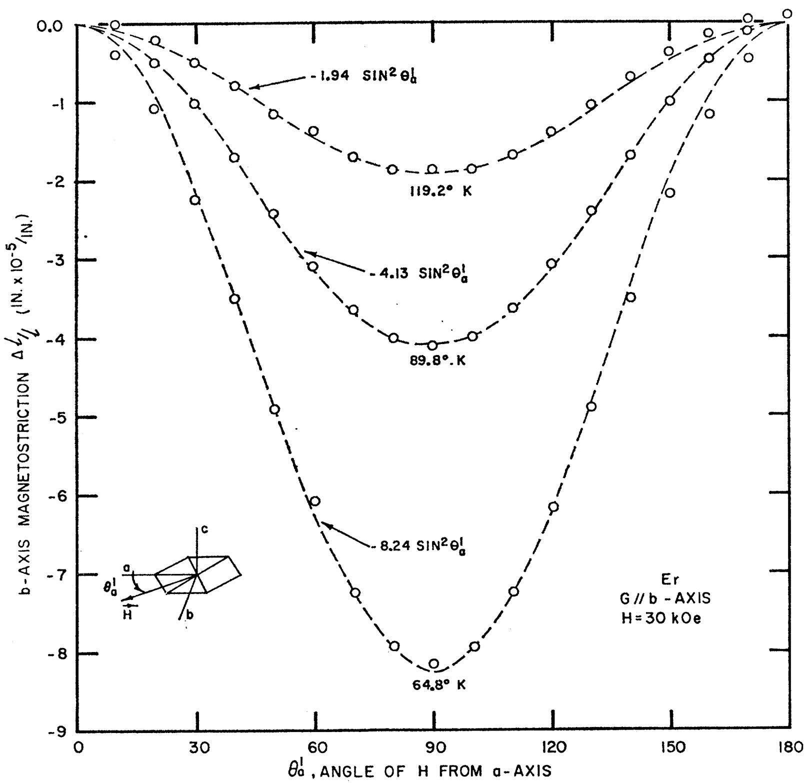

FIG. 8. Second-order theoretical fit and experimental results for the b-axis magnetostriction of Er as a function of the applied field angle relative to the a axis. The 30-kOe field was applied in the basal plane. At 65°K in zero applied field, Er has the modulated-moment structure. At the other temperatures Er is paramagnetic.

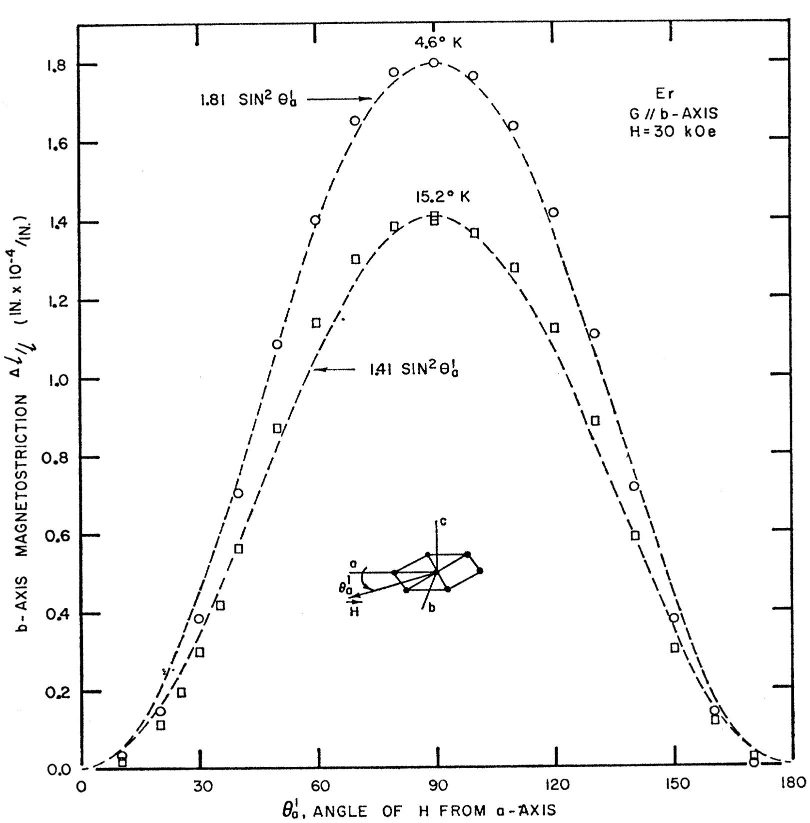

FIG. 9. Second-order theoretical fit and experimental results for the b-axis magnetostriction of Er as a function of applied field angle relative to the a axis. The 30-kOe field was applied in the basal plane. The zero-external-field state is one of conical ferromagnetism at these temperatures.

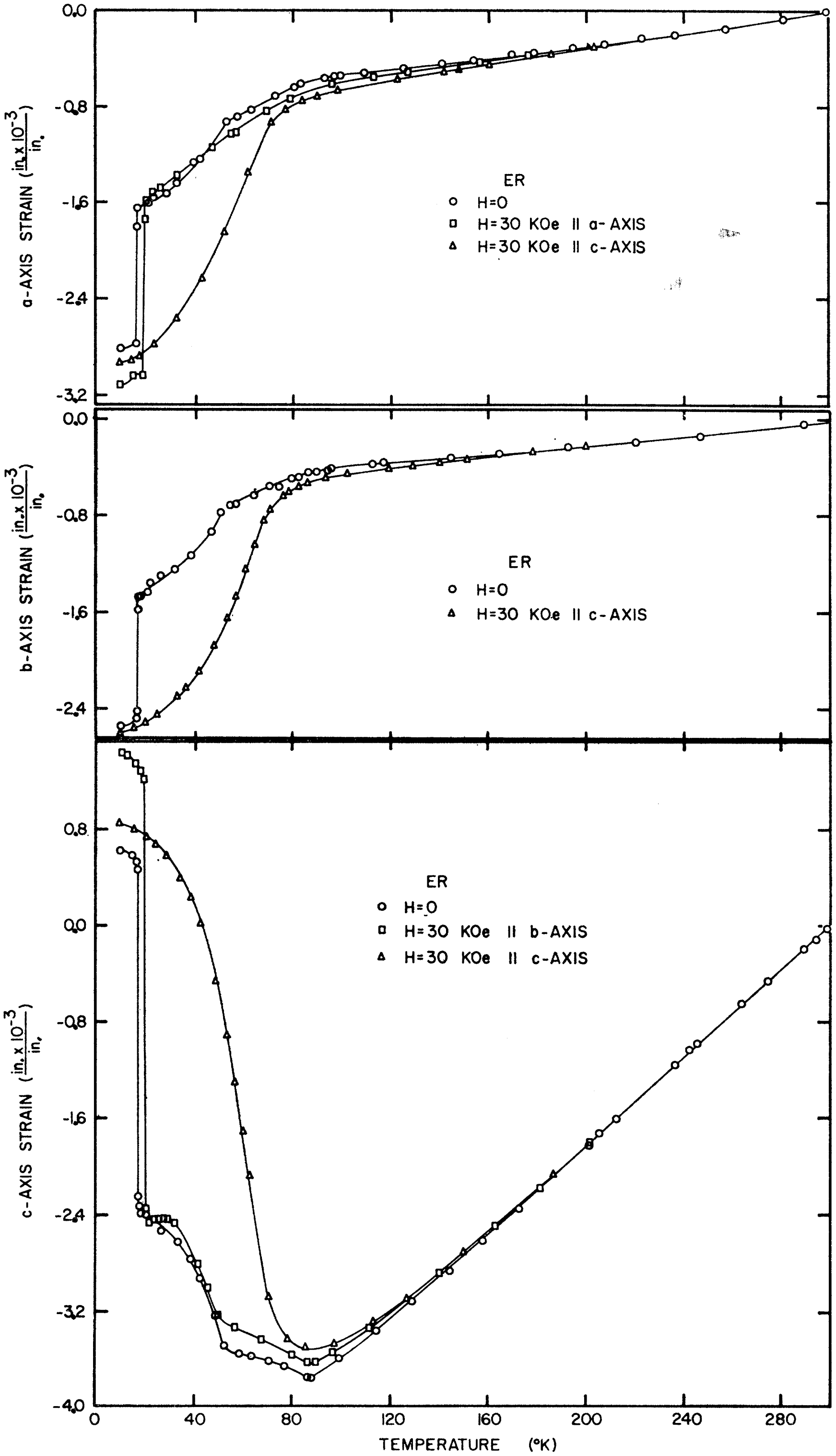

FIG. 10. The a-, b-, and c-axis strain of Er as a function of temperature in zero field and in a 30-kOe field applied along the c axis. Results are also given for the a- and c-axis strains with the field applied in the basal plane.

ment regions where $\phi=90°$. The close agreement to the $\sin^2\theta$ curves indicates the sufficiency of the second-order theory at these temperatures. The strain at $\theta_a'=90°$ is equal to the constant $\lambda^{\gamma,2}$. The values of $\lambda^{\gamma,2}$ are pro portional to H^2 as expected for paramagnetic behavior.

The negative sign of the values indicated for $\lambda^{\gamma,2}$ is opposite to that obtained for the other heavy rare earths and is in agreement with a theory of Tsuya, Clark, and Bozorth.[9] They have calculated the one-ion magnetoelastic coupling coefficients in the rare earths, considering the effects of the distortion of the crystal field by the nearest-neighbor positive ions and the field arising from the strained charge distribution of the conduction electrons. Their theory predicts negative values for the constant $\lambda^{\gamma,2}$ in Er due to the prolate form of the $4f$ electron charge cloud in contrast to the oblate nature of the distribution in Dy, Tb, and Ho which gives rise to a positive magnetostriction.

Callen and Callen[7] have shown on the basis of the one-ion theory that the temperature dependence of the second-order magnetostriction constants is represented by

$$\lambda^{\gamma,2}(T)=\lambda^{\gamma,2}(T=0)I_{5/2}(\mathcal{L}^{-1}(m_n))/I_{1/2}(\mathcal{L}^{-1}(m_n)), \qquad (3)$$

where I is a modified Bessel function of the second kind, $\mathcal{L}^{-1}$ is the inverse Langevin function, and m_n is the reduced magnetization. This expression accurately represents the temperature dependence of $\lambda^{\gamma,2}$ in Tb [2] and Dy.[8] For Er, using the values of $\lambda^{\gamma,2}$ from the angle-dependent magnetostriction data and the magnetic-moment data of Green *et al.*, the anticipated saturation value of $\lambda^{\gamma,2}$ at $T=0$ was calculated from Eq. (3). A value of $\lambda^{\gamma,2}(T=0)=-5.4\times10^{-3}$ in./in. was obtained.

Direct measurement of the b-axis strain as a function of applied field angle θ_a' in the conical ferromagnetic region below 18°K produced the results shown in Fig. 9. Assuming a cone angle $\theta=30°$ the values obtained for $\lambda^{\gamma,2}$ from these data are $+7.24\times10^{-4}$ in./in. and 5.64×10^{-4} in./in. at 4.6 and 15°K, respectively. A possible explanation for the opposite sign and smaller magnitude of the measured strains lies in the neglect of the effects of the exchange magnetostriction in using the one-ion theory to predict the temperature dependence. As discussed earlier, the strain dependence of the exchange energy is the dominant magnetostriction mechanism below 19°K.

In the quasi-antiphase-domain region (18°K$<T<$53°K), the magnitude and the sign of $\lambda^{\gamma,2}$ is a function of applied field value, as can be seen from inspection of the linear magnetostriction curves of Fig. 5. The high-field values of $\lambda^{\gamma,2}$ are negative in this region.

[9] N. Tsuya, A. E. Clark, and R. M. Bozorth, Proceedings of the Nottingham Conference on Magnetism, 1964 (unpublished).

V. TEMPERATURE DEPENDENCE OF Er STRAIN

Figure 10 shows the temperature dependence of the a-, b-, and c-axis strains as a function of temperature. Curves are shown for zero field and for a 30-kOe field applied along the c axis. The a- and c-axis curves also show the strain resulting from application of a 30-kOe field along the a axis. The c-axis field curves show the effects of the field-induced change in the exchange energy below 85°K. Fields applied in the basal plane produce only small modifications of the zero-field strain except below 18°K, where the conical ferromagnetic state is broken down into the fan structure. The zero-field curves show inflection points at about 55 and 85°K corresponding to changes in the states of magnetic order. At the lowest transition temperature, large discontinuities are observed in the zero-field strain corresponding to the change in exchange energy as the axial anisotropy annihilates the quasi-antiphase domain in favor of the conical ferromagnetic structure. As opposed to Dy and Tb, this change is not accompanied by an orthorhombic distortion of the hexagonal lattice, as shown from the x-ray studies of Darnell.[10] The conical ferromagnetic transition occurs spontaneously at 17.5°K and can be induced by a 30-kOe field applied in the basal plane at 20°K.

In principle, the symmetry preserving magnetostriction constants (α modes) can be determined from the isofield data of Fig. 10 as was done for Tb.[2] This was not attempted in Er because of the uncertain nature of the field-induced modifications of the magnetic states.

ACKNOWLEDGMENTS

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[10] F. J. Darnell, Phys. Rev. 132, 1098 (1963).