Magnetoelastic Behavior of Europium Oxide. II. Magnetostriction and the λ Anomaly

B. E. ARGYLE AND N. MIYATA*
IBM Watson Research Center, Yorktown Heights, New York
(Received 5 October 1967)

Magnetostriction of single-crystal EuO was determined in the temperature range 4.2–150°K in applied magnetic fields up to 20 kOe. Linear magnetostriction coefficients extrapolated to 0°K are $\lambda_{100}=-22\times10^{-6}$ and $\lambda_{111}=55\times10^{-6}$. These yield the magnetoelastic coupling constants $b_1(0)\equiv-\frac{3}{2}\lambda_{100}(c_{11}-c_{12})=(31\pm6)\times10^6$ and $b_2(0)\equiv-3\lambda_{111}c_{44}=-(86\pm10)\times10^6$ dyn/cm². Their decrease with increasing temperature appears to be explained by a 1:1 admixture of longitudinal single-ion and two-ion spin correlation as described by $\lambda(T)=\lambda(0)[\hat{I}_{5/2}(\mathfrak{L}^{-1}(m))]$ and $\lambda(T)=\lambda(0)m^2$, respectively. This suggests that the magnetoelastic Hamiltonian contains one-ion (B_1, B_2) and two-ion (D_1, D_2) magnetoelastic constants, $B_1\approx D_1\approx\frac{1}{2}b_1$ and $B_2\approx D_2\approx\frac{1}{2}b_2$. Since classical dipole-dipole interactions in EuO theoretically give $D_1{}^d=-6.4\times10^6$ and $D_2{}^d\approx+4.3\times10^6$, we estimate for pseudodipolar effects $D_1{}^p\approx21\times10^6$ and $D_2{}^p=-47\times10^6$ dyn/cm². The *forced* (volume) magnetostriction λ_f was obtained versus applied magnetic field and is plotted versus the internal magnetic field. These results, which reflect the behavior of short-range order (isotropic spin-spin correlations) in the presence of a magnetic field, are qualitatively similar to predictions of the two-particle cluster calculation by E. R. Callen and H. B. Callen. The Landau-Belov phenomenological theory, which is often used to estimate the pressure derivative of the transition temperature from data of λ_f versus σ^2, leads to results which are inconsistent with other determinations for dT_c/dP in EuO and with the theoretical and experimental results in a previous paper, showing that EuO exhibits a temperature-independent magnetic Grüneisen parameter. Data exhibiting suppression of the λ anomaly in thermal expansivity by applied magnetic fields is also presented. The peak is rounded and its magnitude reduced by a factor of $\approx\frac{1}{2}$ at 18 kOe.

INTRODUCTION

THIS paper presents experimental data of the magnetoelastic strains induced in single-crystal EuO by applied magnetic fields up to 20 kOe at temperatures ranging from 4.2°K to twice the Curie temperature ($T_c=69.2$°K). The anisotropic and isotropic components of magnetostrain (linear and volume magnetostrictions, respectively) are determined, and they are used to characterize the magnetoelastic interactions which presumably arise from single-ion and two-ion terms in a microscopic theory as formulated by E. Callen and H. B. Callen.[1]

The *spontaneous* magnetostrain, which is obtainable from the area under the λ anomaly in the thermal expansivity, was previously investigated by Argyle, Miyata, and Schultz[2] (hereafter referred to as I). This effect results from coupling between elastic and magnetic degrees of freedom through the volume dependence of the two-ion, isotropic exchange interactions, $-2\sum J_{ij}(V)\mathbf{S}_i\cdot\mathbf{S}_j$. The advantage of studying EuO in contrast to other members of the chalcogenide series, EuS, EuSe, and EuTe, is the apparent predominance of nearest-neighbor-type interactions.[3] The strength of the exchange magnetoelastic coupling in EuO ($\gamma_m\equiv\partial\ln J/\partial\ln V=-5.3$) was determined in I from the magnetic Grüneisen relation $\alpha_{\rm me}=-\gamma_m KC_m/3V$ by comparing the magnetoelastic component of expansivity, $\alpha_{\rm me}\equiv(d/dT)(\delta l/l_0)_{\rm me}$, with the magnetic specific heat $C_m\equiv(dU_m/dT)_{H,V}$. These magnetic quantities were obtained by correcting the observed expansivity and specific heat for estimated contributions of the lattice. The coupling coefficient γ_m was observed to be temperature-independent and may thus be termed a "*magnetic* Grüneisen constant" in analogy with the lattice Grüneisen constant which occurs in the standard theory of lattice expansivity.

From theoretical considerations we have shown also in I that the width of the region at $T\cong T_c$ where the proportionality relation between $\alpha_{\rm me}$ and C_m must break down may be so narrow in EuO as to be undetectable. Further, the changes in J with temperature caused by the total thermal strain $[\delta J/J=\gamma_m\int_0{}^T 3\alpha_{\rm obs}(T)\,dT]$ are less than 3% over the temperature range of this experiment (see Table I of Ref. 2). Thus in EuO the anomalous expansivity may be utilized to measure the temperature variation of the internal magnetic energy which in turn reflects correlations of spin-pairs coupled by the short-range exchange forces.

While specific-heat measurements give this information from a more fundamental standpoint, they are difficult to obtain for $H>0$. We are interested then in extending our measurements to the realm of "forced" magnetostrain in order to reveal the behavior of spin-pair correlation when $H>0$ and to exhibit the suppression of the λ anomaly by a magnetic field. These results obtained on this material are particularly meaningful

* Present address: Yokohama National University, Minamiku, Yokohama, Japan.

[1] (a) E. Callen and H. B. Callen, Phys. Rev. **129**, 578 (1963); (b) **139**, A455 (1965).

[2] B. E. Argyle, N. Miyata, and T. D. Schultz, Phys. Rev. **160**, 413 (1967).

[3] T. R. McGuire, B. E. Argyle, M. W. Shafer, and J. S. Smart, J. Appl. Phys. **34**, 1345 (1963).

to compare with theoretical calculations[4] based on the Heisenberg Hamiltonian because the necessary approximations and complexities in the theory are minimized for the case of a cubic, single sublattice ferromagnet with S-state ions [in this case, $Eu^{++}(^8S_{7/2})$]. Also, among the numerous experimental attempts[5] to characterize the nature of the second-order phase transformation, our approach using expansivity and magnetostriction data is unique.

Before comparing our data with the isotropic theory, the usual anisotropic effects, namely, magnetocrystalline anisotropy (K_1) and linear magnetostriction (λ_{100}, λ_{111}), need to be accounted for or shown to be small. Magnetocrystalline anisotropy, usually described by $K_1\sum_{i>j}\alpha_i^2\alpha_j^2$ in the free energy, may include effects of anisotropic exchange and/or single-ion interactions with the crystal potential. We have observed[6] that the variation of K_1 with the magnetization $\sigma(H,T)$ is remarkably well described by predictions for a single-ion mechanism.[7,8] This suggests that the dominant origin of anisotropy is the splitting of $^8S_{7/2}$ levels of the $4f$ electrons by the octahedral crystal field. Its strength in EuO is specified by the cubic crystal-field splitting parameter $b_4=21.2\times10^{-4}$ cm^{-1}/ion.[6] This is two orders of magnitude smaller than the exchange splitting which is characterized by the Heisenberg exchange coefficient $J/k\approx0.75$°K or 0.5 cm^{-1}/(ion pair).[9] Similarly, the elastic energy associated with the anisotropic (linear) magnetostrain reported here in EuO is two orders of magnitude smaller than the magnetocrystalline anisotropy energy. Thus we may expect negligible contribution of these anisotropic effects to the observables (e.g., magnetic energy, magnetic specific heat, and magnetoelastic expansivity) that are basic to a comparison with the isotropic theory.

The linear magnetostriction of a cubic ferromagnet arises from coupling between the elastic and magnetic anisotropy energies.[10] The microscopic origins can be classified as to the number of ions involved; E. and H. B. Callen derived the behavior arising from single-ion[1a] and two-ion[1b] energies while neglecting inhomogeneous or phonon modes of lattice strain. The temperature variation is described by appropriate (longitudinal) single-ion and pair correlations provided that the magnetoelastic coupling strength is temperature-independent. That this approximation may be reasonable for EuO is suggested both by its having rather low compressibility ($K_T\approx10^{-12}$ dyn^{-1} cm^2) and by an evidently small variation of compressibility with temperature ($\Delta K/K\Delta T\approx10^{-5}$).[2] We shall investigate the question of the relative strength of one- and two-ion magnetostrictive coupling by comparing our data with the Callen theory[1] for the dependence of longitudinal single-ion and two-ion spin correlations on the magnetization. It might be expected that mainly single-ion effects occur in linear magnetostriction of EuO since the magnetic anisotropy energy itself appears to obey the one-ion theory.[6]

Phenomenological Equation of Magnetostriction

Europium oxide has the cubic NaCl crystal structure and for this type of symmetry the homogeneous magnetoelastic strain $(\delta l/l)_{\rm me}$ may be represented by the expression

$$(\delta l/l_0)_{\rm me}=\lambda_{\rm vol}+\lambda_{\rm lin} \tag{1}$$

with the separate strain modes being completely independent ($\lambda_{\rm vol}$) of, or dependent ($\lambda_{\rm lin}$) on the direction of observation of the strain. The predominant effect, the spontaneous *volume* strain λ_S associated with the expansion anomaly, was investigated in I. Also of interest are the dilatations observed on applying a magnetic field. Up to terms believed significant to the accuracy of our measurements, these magnetostrains may be described as follows:

$$\lambda_{\rm vol}=\lambda_f+\lambda_c\sum_{i,j}(\alpha_i^2\alpha_j^2-\tfrac{1}{3})+\lambda_N, \tag{2}$$

$$\lambda_{\rm lin}=\tfrac{3}{2}\lambda_{100}\sum_i(\alpha_i^2\beta_i^2-\tfrac{1}{3})+3\lambda_{111}\sum_{i,j}\alpha_i\alpha_j\beta_i\beta_j, \tag{3}$$

where α_i and β_i denote direction cosines with respect to the crystal axes of the magnetization and observation direction, respectively. [A procedure for extracting the λ coefficients by observing $(\delta l/l)_{\rm me}$ versus H is described in the next section.] Other terms of higher order in α_i^2 may exist, but their strength in other ferromagnets is usually undetectable. The second-order term (λ_c) in Eq. (2) is also usually neglected, but it may be argued that when the applied fields can scan up to a significant fraction of the Weiss field, this term may be observable.

The nature and phenomenological origin of these terms may be described as follows: While $\lambda_{\rm vol}$ is that part of the total strain which is symmetric, its amplitude may be modulated, however, when the magnetization is rotated against the torques of magnetocrystalline anisotropy. This effect is described by $\lambda_c\sum_{i,j}(\alpha_i^2\alpha_j^2-\frac{1}{3})$ and has been called the "crystal effect," while that part in Eq. (2) which is independent of α_i is the isotropic forced magnetostriction λ_f. Forced magnetostriction relates to an increase in spin alignment induced by a magnetic field and has the same origin as the spontaneous magnetic strain (described in I) which occurs on cooling the specimen through the transition temperature in zero field. Because the demagnetization energy depends on the volume of the specimen, an additional volume strain λ_N is included in Eq. (1).

[4] H. B. Callen and Earl R. Callen, Phys. Rev. **130,** 890 (1963); **136,** A1675 (1964); J. A. Copeland and H. A. Gersch, *ibid.* **143,** 236 (1966); G. S. Rushbrook and P. J. Wood, Mol. Phys. **1,** 257 (1958); also Ref. 1(b).

[5] See, e.g., Nat. Bur. Std. (U.S.) Misc. Publ. **273** (1966); C. Domb, *Low Temperature Transitions* (Plenum Press, Inc., New York, 1965), Vol. 9, Part B.

[6] N. Miyata and B. E. Argyle, Phys. Rev. **157,** 448 (1967).

[7] W. P. Wolf, Phys. Rev. **108,** 1152 (1957).

[8] H. B. Callen and S. Shtrikman, Solid State Commun. **3,** 5 (1965).

[9] E. L. Boyd, Phys. Rev. **145,** 174 (1966).

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

This "form effect"[11] saturates after technical saturation of the magnetization. Thus we shall be able to distinguish between λ_N and λ_f because they vary quite differently with H and T. The linear magnetostriction λ_{lin} depends on both α_i and β_i and is written in Eq. (3) so as to omit any change of volume. Here we use a two-constant expression that represents a change of length encountered between the demagnetized state, represented as an equilibrium distribution of domain orientations along [111] easy axes of magnetization, and the magnetized state. The constants λ_{111} and λ_{100}, called "linear magnetostriction coefficients," may contain both spontaneous and induced effects. The spontaneous effect is observed only after applying a field sufficient to destroy the demagnetized state.

Microscopic Theory

While the equations (1)–(3) of magnetostriction extended to include terms to fourth degree in α_i were originally derived by Becker and Döring[12] from a phenomenological free energy restricted by crystal symmetry, E. and H. B. Callen[1] have from a microscopic magnetoelastic Hamiltonian using group theory shown that equivalent results may be obtained quantum mechanically. The important new result of E. and H. B. Callen treatment is that the λ coefficients are expressed in terms of certain averages of spin functions whose temperature variation depends on the kind of magnetic interaction.

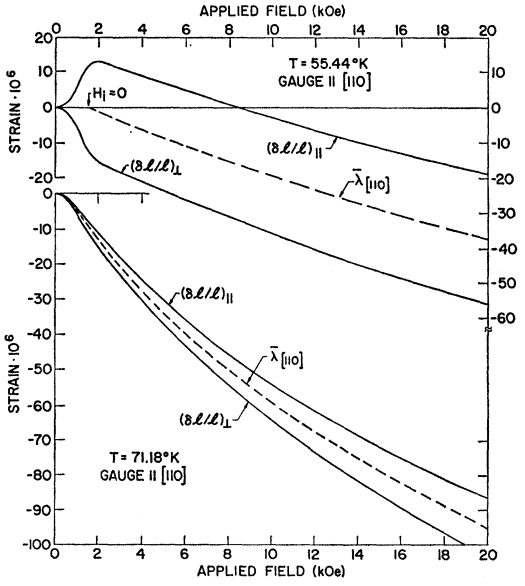

FIG. 1. Solid curves are examples of strain $\delta l/l$ versus magnetic field applied parallel $(\delta l/l)_{||}$ and perpendicular $(\delta l/l)_{\perp}$ to gauge orientation [110] in (100) plane for temperatures above (lower plot) and below (upper plot) the Curie temperature (T_c=69.2°K) of EuO. Dotted curves are mean values of $\lambda_{||}$ and $\lambda_{\perp}$.

[11] S. Chikazumi, *Physics of Magnetism* (John Wiley & Sons, Inc., New York, 1964), Chap. 8.

[12] R. Becker and W. Döring, *Ferromagnetism* (Julius Springer-Verlag, Berlin, 1939).

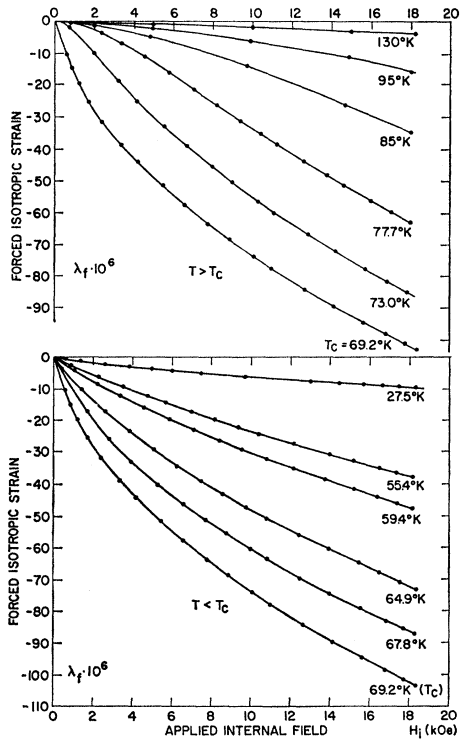

FIG. 2. Isotropic component of induced strain λ_f (forced magnetostrain) versus internal magnetic field H_i applied at several temperatures above (upper plot) and below (lower plot) the Curie temperature.

E. and H. B. Callen studied in detail the case when one-ion[1a] and two-ion[1b] magnetoelastic interactions are treated as perturbation terms in a Hamiltonian containing the elastic energy and the isotropic and anisotropic magnetic energies. (The magnetoelastic terms arise from the strain dependence of the magnetic energies.) The 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 β is given by $\delta l/l = \sum_{i,j} e_{ij}\beta_i\beta_j$, where e_{ij} are the equilibrium magnetostrains. The resulting expression has the form of Eqs. (1)–(3) except that the isotropic strain λ_{vol} in the Callen theory includes the spontaneous magnetostriction λ_s as well as λ_f and omits the crystal effect λ_c and the form effect λ_N which necessarily occur in a real crystal. In order to compare our data on EuO with theory, we are required to extract separately λ_s, λ_f, λ_{111}, and λ_{100} from the observed strains. Our procedure to achieve this is given below.

SAMPLE AND EXPERIMENTAL METHOD

The single-crystal specimen and experimental technique used to observe strain $\delta l/l$ are the same as given in I. Two wire strain gauges (Kyowa-type 19-1S2) are

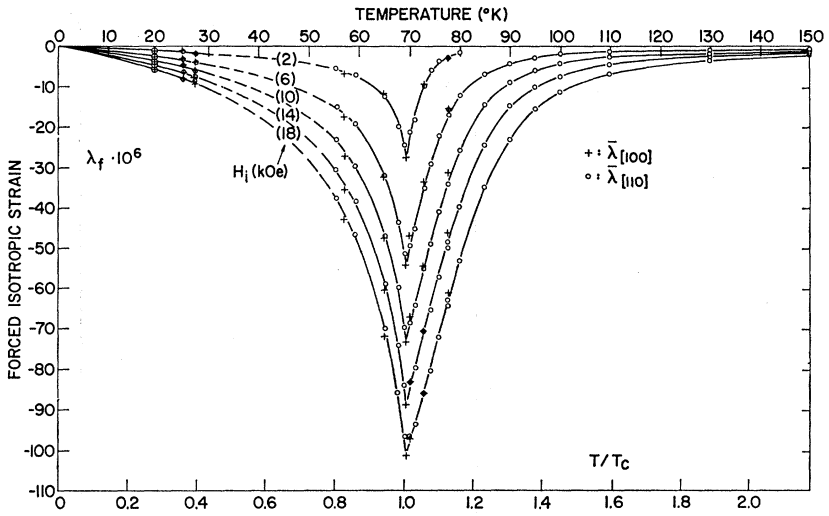

FIG. 3. Isotropic forced magnetostrain (λ_f) versus temperature at various fixed internal applied magnetic fields. Data represented by circles and crosses were obtained from strain gauges oriented along ([110], [100]) crystal axes.

attached to an (001)-disk specimen parallel to [100] or [111] axes. One of these active gauges and three reference gauges attached to copper plates comprise a Wheatstone-bridge arrangement. By special arrangement, changes in the unbalance voltage of the bridge are quite accurately proportional to the difference between the strains of the EuO crystal and the nonmagnetic copper. It is well known that the magnetic field and temperature dependence of the gauge resistance R and strain gauge factor $G \equiv (\delta R/R)/(\delta l/l)$ may destroy this proportionality. Gersdorf[13] has measured the properties of these gauges in detail. The magnetoresistivity effects that he observed (both isotropic and anisotropic with respect to field orientation) are small and were nearly cancelled in our arrangement as follows: Two reference gauges connected in electrically adjacent arms of the bridge are positioned with their axes perpendicular to the applied field. The third reference gauge is positioned parallel to the active gauge which is fixed in the plane of rotation of the applied field, and this pair is also connected adjacent to each other in the Wheatstone-bridge circuit. Thus while a field of fixed magnitude is rotated, the anisotropic contribution from the magnetoresistivity of the gauges does not contribute to the bridge's unbalance. Similarly, when the magnitude of the field (with fixed orientation) is changed, that part of the gauge magnetoresistivity dependent on the magnitude of the field is also compensated. The change in bridge unbalance is therefore proportional to the changes in the magnetostrain of the specimen. From Gersdorf's study of Kyowa gauges, the ratio R/G of gauge resistance to gauge factor is nearly constant (within 2%) in the temperature range of our experiment. Thus our calibration, which is based on this ratio, was determined at 77°K and is here treated as a constant.

[13] R. Gersdorf, Ph.D. thesis, University of Amsterdam, 1962 (unpublished).

The strain $\delta l/l$, observed after applying the field in a (001) plane at an angle θ from the [100] direction, was analyzed in terms of Eqs. (1)–(3). These expressions simplify as follows when the field is parallel or perpendicular to the strain gauge:

$H \parallel$ [100] gauge:

$$(\delta l/l)_{[100],[100]} = \lambda_f + \lambda_N + \lambda_{100}, \tag{4}$$

$H \perp$ [100] gauge:

$$(\delta l/l)_{[100],[010]} = \lambda_f + \lambda_N - \tfrac{1}{2}\lambda_{100}, \tag{5}$$

$H \parallel$ [110] gauge:

$$(\delta l/l)_{[110],[110]} = \lambda_f + \tfrac{1}{4}\lambda_c + \lambda_N + \tfrac{1}{4}\lambda_{100} + \tfrac{3}{4}\lambda_{111}, \tag{6}$$

$H \perp$ [110] gauge:

$$(\delta l/l)_{[110],[1\bar{1}0]} = \lambda_f + \tfrac{1}{4}\lambda_c + \lambda_N + \tfrac{1}{4}\lambda_{100} - \tfrac{3}{4}\lambda_{111}. \tag{7}$$

Considering Eqs. (4)–(7) as four equations in four unknowns, the quantity $\lambda_f + \lambda_N$ being one unknown, we obtain the λ coefficients by inverting these equations as follows:

$$\lambda_{100} = \tfrac{2}{3}\{(\delta l/l)_{[100],[100]} - (\delta l/l)_{[100],[010]}\}, \tag{8}$$

$$\lambda_{111} = \tfrac{2}{3}\{(\delta l/l)_{[110],[110]} - (\delta l/l)_{[110],[1\bar{1}0]}\}, \tag{9}$$

$$\lambda_f + \lambda_N = \tfrac{1}{3}\{(\delta l/l)_{[100],[100]} + 2(\delta l/l)_{[100],[010]}\} \equiv \bar{\lambda}_{[100]}, \tag{10}$$

$$\lambda_f + \tfrac{1}{4}\lambda_c + \lambda_N + \tfrac{1}{4}\lambda_{100} = \tfrac{1}{2}\{(\delta l/l)_{[110],[110]} + (\delta l/l)_{[110],[1\bar{1}0]}\} \equiv \bar{\lambda}_{[110]}, \tag{11}$$

where we have also introduced the averages $\bar{\lambda}_{[100]}$ and $\bar{\lambda}_{[110]}$ of the observed strains for $H_{\parallel}$ and $H_{\perp}$ weighted according to Eqs. (10) and (11).

As suggested by the form of Eqs. (8)–(11), the temperature was held fixed (to within $\approx$0.01°K) and the strain was either measured by applying $H_{\parallel}$ and $H_{\perp}$ or by rotating H in the (001) plane. Examples of data resulting from the first case obtained using the [110] gauge are given in Fig. 1. The upper plot for 55.44°K

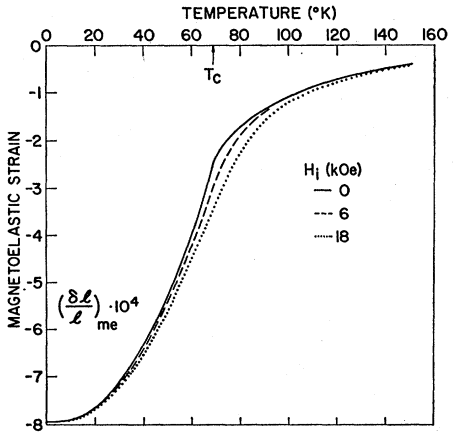

FIG. 4. Isotropic magnetoelastic thermal strain $(\delta l/l)_{me}$ versus temperature at various fixed internal magnetic fields H_i. Curve for $H_i=0$ is spontaneous magnetoelastic strain obtained in I (Ref. 2).

(i.e., less than T_c) exhibits a superposition of spontaneous and induced strain. The spontaneous strain manifests itself by the "knee" which appears near the field required to overcome the demagnetizing field (internal field, $H_i=0$). In the lower plot for $T=71.18°K>T_c$, the spontaneous strain is zero. In each case the average of the two solid curves (for $H_{||}$ and $H_\perp$) is shown as the dashed curve and this gives $\bar{\lambda}_{[110]}=\lambda_f+\frac{1}{4}\lambda_c+\lambda_N+\frac{1}{4}\lambda_{100}$ according to Eq. (11). Data similarly observed using the [100] gauge give $\lambda_f+\lambda_N\equiv\bar{\lambda}_{[100]}$ from the (weighted) average in Eq. (10). To evaluate λ_f we must compensate for $\frac{1}{4}\lambda_{100}$, λ_N, and $\frac{1}{4}\lambda_c$ in the curves of $\bar{\lambda}_{[110]}$ versus H, and correct for λ_N in the curves of $\bar{\lambda}_{[100]}$ versus H.

The form effect λ_N may be estimated from the relation[11]

$$\lambda_N(0)=\tfrac{1}{3}NM_0^2/2B=0.8\times10^{-6}$$

by taking N (demagnetizing factor) $=1.45$, M_0 (magnetization at 0°K) $=1.9\times10^3$ emu/cm³, and B (bulk modulus) $=1.07\times10^{12}$ dyn/cm². A strain of this magnitude was observed at the initial part of the magnetization for temperatures below ~30°K. But

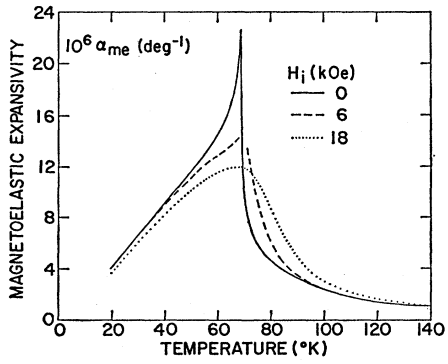

FIG. 5. Magnetoelastic thermal expansivity $\alpha_{me}\equiv(d/dT)(\delta l/l)_{me}$ versus temperature at various fixed internal magnetic fields H_i. Curve for $H_i=0$ is from I (Ref. 2).

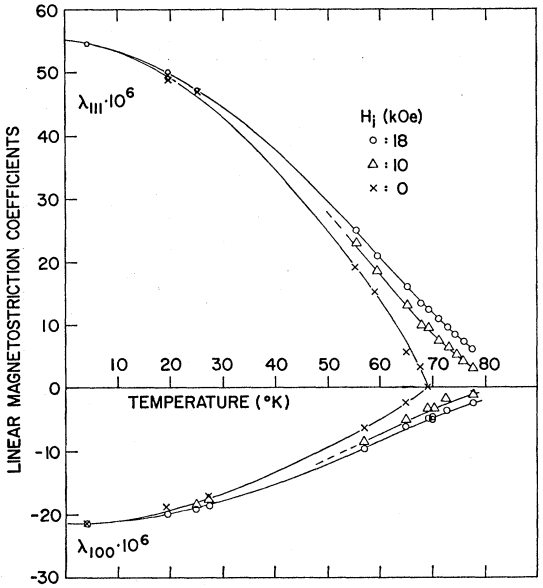

FIG. 6. Linear magnetostriction coefficients (λ_{100}, λ_{111}) versus temperature at various fixed internal magnetic fields.

for evaluating λ_f at these temperatures, we restrict our attention to fields above saturation and therefore omit λ_N by extrapolation in this region where the magnetization is nearly independent of H. On the other hand, at temperatures near T_c the forced magnetization is comparable to or larger than the spontaneous magnetization. In this region we estimate $\lambda_N\leq0.2\times10^{-6}$ since λ_N is proportional to M^2. This is negligible compared with $\lambda_f\cong5\times10^{-5}$.

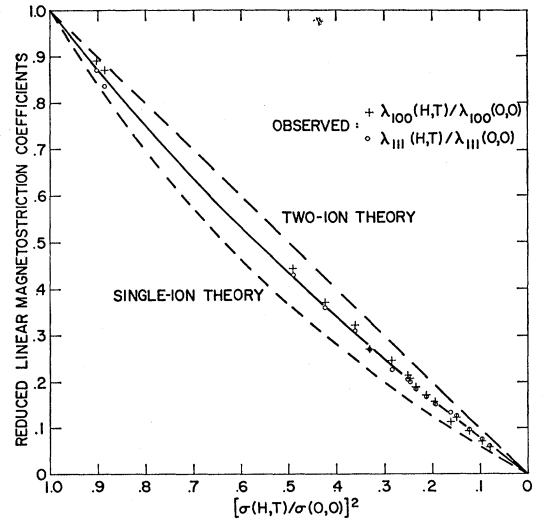

FIG. 7. Plus signs and circles, reduced linear magnetostriction coefficients versus magnetization at various temperatures and magnetic fields. Two dashed curves, theoretical predictions by E. R. Callen and H. B. Callen (Ref. 1) based on one- and two-spin cluster theory (see text). Solid curve, linear combination of the two theoretical curves taken with equal weight.

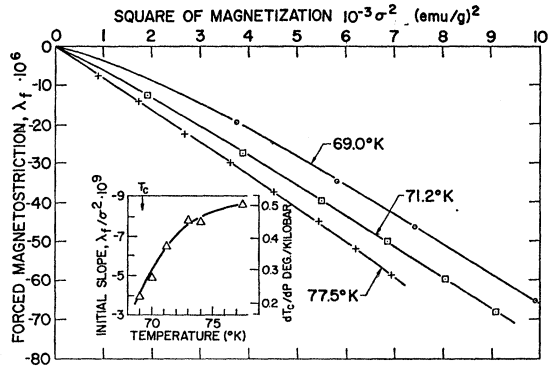

FIG. 8. Isotropic forced magnetostrain (λ_f) versus magnetization squared for fixed temperatures near $T_c=69.2$°K. Inset gives values of dT_c/dP derived from initial slopes using Landau-Belov theory. See test for values given by other experimenters.

The λ_{100} coefficient was determined as a function of **H** by taking the difference between data curves for **H** oriented parallel and perpendicular to the [100] gauge [Eq. (8)]. Similarly, λ_{111} was determined using the [110] gauge and Eq. (9). The results, $\lambda_{100}(T,H_i)$ and $\lambda_{111}(T,H_i)$, are plotted in Fig. 6 versus T for various fixed values of *internal* magnetic field H_i determined from the magnetization behavior (σ versus H) and demagnetization factor of the specimen.

In the determination of $\lambda_f(H,T)$ only the induced part of $\frac{1}{4}\lambda_{100}$ need be compensated for. We find this to be 1–2% of λ_f when determined at the same magnetic field and temperature.

The correction for the crystal effect $(\frac{1}{4}\lambda_c)_{\text{ind}}$, quite fortunately, has the same magnitude and opposite sign as $(\frac{1}{4}\lambda_{100})_{\text{ind}}$ as found by making comparison between $\bar{\lambda}_{[100]}$ and $\bar{\lambda}_{[110]}-\frac{1}{4}(\lambda_{100})_{\text{ind}}$. Therefore, with cancellation of these two terms in Eq. (11), the magnetic-field dependence of λ_f at fixed temperatures shown in Fig. 2 was obtained directly from data for $H_{||}$ and $H_\perp$ by the weighted averages $\bar{\lambda}_{[100]}$ and $\bar{\lambda}_{[110]}$ of Eqs. (10) and (11)—thus omitting the induced part of $\frac{1}{4}\lambda_c$, $\frac{1}{4}\lambda_{100}$, and λ_N.

EXPERIMENTAL RESULTS

Figures 2–5 reveal quantitative details of the effect of a magnetic field on the magnetic phase transition in EuO while omitting anisotropic effects. Figure 2 presents data of λ_f versus internal magnetic field H_i. The temperature dependence of λ_f for constant values of H_i given in Fig. 3 is obtainable directly from Fig. 2. The success of the above procedure of correcting for anisotropic effects is amply demonstrated by the equivalent results for λ_f (circles and crosses) obtained from different strain gauges ([110] and [100] orientations). The temperature dependence of the total isotropic magnetoelastic strain $(\delta l/l)_{\text{me}}$ for fixed values of H_i is shown in Fig. 4. For $H=0$ the curve was obtained by integrating the area under the expansivity anomaly obtained in I and also shown in Fig. 5, while the curves for $H_i=6$ and 18 kOe were obtained by combining the data in Fig. 3 with the curve for $H_i=0$. Similarly, by combining the slope $d\lambda_f/dT$ of the curves in Fig. 3 with data of the expansivity coefficient α_{me} ($H=0$) obtained in I, it is shown in Fig. 5 how the peak in the expansivity anomaly is decreased and broadened by application of a magnetic field. Viewed on a coarse scale, the position of the peak appears unaffected by the field. A field of 18 kOe reduces the peak value to $\approx\frac{1}{2}$ the value observed at $H_i=0$ although the latter may be limited by the resolution of the experiment. From the observed proportionality between expansivity and specific heat, we may expect a similar suppression of the λ anomaly in the magnetic specific heat.

The linear magnetostriction constants λ_{100} and λ_{111} are given in Fig. 6 as a function of temperature for fixed magnetic fields. Under all conditions of T and H we observe that $\lambda_{111}/\lambda_{100}=-2.55\pm0.1$. The values extrapolated to absolute zero are $\lambda_{100}(0)=-22\times10^{-6}$ and $\lambda_{111}(0)=55\times10^{-6}$. [Note that these anisotropic strains are small by comparison with the zero-temperature *volume* deformation $\delta_0\equiv(\delta V/V_0)_{\text{me}}=-0.24\%$ given by three times the magnetoelastic strain $(\delta l/l_0)_{\text{me}}$ at $T=0$°K (Fig. 4).] For the usual magnetoelastic coefficients given by the relations[11]

$$b_0=-\tfrac{1}{3}(\delta V/V_0)(c_{11}+2c_{12}),$$

$$b_1=-\tfrac{3}{2}\lambda_{100}(c_{11}-c_{12}),$$

and

$$b_2=-\lambda_{111}3c_{44},$$

we obtain $b_0(0)=2.6\times10^{11}$, $b_1(0)=(31\pm6)\times10^6$, and $b_2(0)=-(86\pm5)\times10^6$ dyn/cm^2 using preliminary elastic constants $c_{11}=(17\pm1)\times10^{11}$, $c_{12}=(7.5\pm1.0)\times10^{11}$, and $c_{44}=(5.2\pm0.6)\times10^{11}$ dyn/cm^2, measured ultrasonically at room temperature.[2] The above signs of b_1 and b_2 are opposite to what may be calculated for purely magnetic dipole-dipole interactions in EuO. This suggests a more complex origin even considering the unusually large magnetic dipole moment ($S=\frac{7}{2}$) of the Eu^{++} ions.

COMPARISON WITH THEORY

Linear Magnetostriction

For EuO, the microscopic theory for λ_{100} and λ_{111} may be summarized as follows:

$$\lambda_{100}=B_1\mathfrak{L}_f(T,H)+D_1\mathfrak{L}_{fg}(T,H), \quad (12)$$

$$\lambda_{111}=B_2\mathfrak{L}_f(T,H)+D_2\mathfrak{L}_{fg}(T,H), \quad (13)$$

where B_1, B_2 and D_1, D_2 are, respectively, one- and two-ion magnetoelastic coupling coefficients (considered to be independent of temperature), and $\mathfrak{L}_f$ and $\mathfrak{L}_{fg}$ are quantum statistical spin averages. These have been called "longitudinal correlation functions," and they are defined in the notation of E. and H. B. Callen[1b]

by

$$\mathfrak{L}_f \equiv \langle (S_f^z)^2 - \tfrac{1}{3}S(S+1) \rangle, \quad (14)$$

$$\mathfrak{L}_{fg} \equiv \langle S_f^z S_g^z - \tfrac{1}{3}(S_f S_g) \rangle. \quad (15)$$

Callen and Shtrikman[8] showed that the one-ion function $\mathfrak{L}_f$ may be expressed as a function of the observed magnetization even though an accurate theoretical expression of the magnetization behavior is unavailable. They obtain

$$\mathfrak{L}_f(T,H)/\mathfrak{L}_f(0,0) = \hat{I}_{5/2}[\mathfrak{L}^{-1}(m)], \quad (16)$$

where $\mathfrak{L}(0,0)$ is the correlation function at $T=0°K$, $\hat{I}_{5/2}$ is a ratio of hyperbolic Bessel functions $(I_{5/2}/I_{1/2})$, and $\mathfrak{L}^{-1}$ is an inverse Langevin function of the reduced magnetization $m \equiv M(T,H)/M_0$. This one-ion theory is plotted versus m^2 in Fig. 7.

The calculation for spin-pair correlations $\mathfrak{L}_{fg}$ depends on the range of interaction. Using the two-particle cluster approximation, E. and H. B. Callen computed $\mathfrak{L}_{fg}(T,H)$, neglecting exchange interactions ranging beyond second-nearest neighbors.[1b] The expression $\mathfrak{L}_{fg}(T,H)/\mathfrak{L}_{fg}(0,0) = m^2$ fit their results (within a few percent) which they computed for several assumed ratios of second-neighbor to nearest-neighbor exchange ranging from -0.4 to 1.0 (see Fig. 8 of Ref. 1b). This expression gives the straight dashed line in Fig. 6.

The data of reduced magnetostriction coefficients, $\lambda_{111}(T,H)/\lambda_{111}(0,0)$, and $\lambda_{100}(T,H)/\lambda_{111}(0,0)$, for EuO (Fig. 7) are explained quite well by a linear combination of the single-ion and two-ion functions weighted equally. This combination is given by the solid line passing through the data points. Thus in EuO the microscopic magnetoelastic coupling constants appearing in Eqs. (12) and (13) are

$$B_1 \approx D_1 \approx \tfrac{1}{2}b_1(0) = (15 \pm 3) \times 10^6 \text{dyn/cm}^2,$$

$$B_2 \approx D_2 \approx \tfrac{1}{2}b_2(0) = -(43 \pm 3) \times 10^6 \text{dyn/cm}^2.$$

These constants can be calculated in principle from knowing details of the interaction. They are related to the strain dependence of the magnetocrystalline anisotropy energy.[11,12] The B coefficients provide information about the strain dependence of the crystal-field potential, but theoretical estimates for EuO are not yet available. Considering two-ion effects, classical magnetic dipole-dipole interactions contribute an amount expressible as[12]

$$D_1^{\text{dip}} = -3SM_s^2, \quad D_2^{\text{dip}} = 2SM_s^2,$$

where M_s is the magnetization and S is a lattice sum. We find using $M_s(0) = 1890$ G and $S = 0.6$ [12] for the fcc lattice of EuO,

$$D_1^{\text{dip}} = -6.4 \times 10^6 \quad \text{and} \quad D_2^{\text{dip}} = 4.3 \times 10^6 \text{dyn/cm}^2.$$

Correcting the observed values $(\frac{1}{2}b_1, \frac{1}{2}b_2)$ by this amount provides an estimate for the quantum-mechanical pseudodipolar contribution

$$D_1^{\text{pseudodipole}} = 21 \times 10^6 \text{dyn/cm}^2,$$

$$D_2^{\text{pseudodipole}} = -47 \times 10^6 \text{dyn/cm}^2.$$

Theoretical estimates for these quantities are not yet available.

Forced Magnetostriction

The data of λ_f versus H_i in Fig. 2 are qualitatively similar to the isotropic spin-pair correlations $\mathcal{J}_{fg}(H,T) = \langle \mathbf{S}_f \cdot \mathbf{S}_g \rangle$ calculated for EuS($T_c = 16.5°K$) by E. and H. B. Callen using the two-particle cluster approximation (see Fig. 6 of Ref. 1b). The region of double concavity appearing in the data curves for temperatures just above T_c (Fig. 2) appears in the theory as well. However, this theory does not explain the λ_f data in detail as we might suspect because, as we have shown in I, it does not quantitatively explain the spontaneous magnetoelastic strain $\lambda_s(T)$.

COMPARISON WITH LANDAU-BELOV THEORY

The Landau phenomenological theory of second-order phase transformations, which is based on the assumption that the free energy is analytic in the neighborhood of the transition, describes thermodynamic properties (i.e., susceptibility, specific heat, etc.) of ferromagnetic systems although not with the best precision. Nevertheless, the Landau-Belov extension[14] to a magnetoelastic system has often been used to predict the pressure dependence of the Curie temperature in metals.[14,15] We may test this idea using EuO as an example of an insulating ferromagnet, and conveniently so, since we have previously determined $dT_c/dP = 0.34 \pm 0.02$ deg/kbar[2] from the magnetic Grüneisen parameter in I.

Belov[14] derived relations for the critical region giving

$$\sigma \propto (T - T_c)^{1/2},$$

$$\sigma \propto H^{1/3},$$

and

$$\lambda_f \propto \sigma^2$$

by expanding the free energy at constant magnetization σ in a power series in σ^2 and $T - T_c$ and by making the assumption that the pressure P appears in the term in σ^2 and appears only linearly. Expressions for the first two proportionalities are equivalent to the predictions of molecular-field theory except that the coefficients are not specified but are rather to be treated as undetermined parameters. Of primary interest to us is the resulting new relation derived by Belov,

$$dT_c/dP = -C_c/A_c, \quad (17)$$

where, using the notation of Nakajima,[15] A_c is a constant given by $A = A_c(T - T_c)$ and A, the coefficient of the σ^2 term in the phenomenological free energy, can be obtained from magnetization versus field data. Also, C_c is given by $6\gamma_c/\rho$, where ρ is the

[14] K. P. Belov, *Magnetic Transitions* (Boston Technical Publishers, Inc., Cambridge, Mass., 1965).
[15] T. Nakajima, J. Phys. Soc. Japan **19**, 520 (1964).

density and γ_c is the constant of proportionality between forced magnetostriction $\lambda_f(H)$ and $\sigma^2(H)$ determined at $T=T_c$.

For analysis of EuO in terms of this theory, data of λ_f are plotted against σ^2 in Fig. 8. We note first that linearity between λ_f and σ^2 does not occur at $T=T_c=$ 69°K as predicted, and secondly that except in the region of low fields, σ is not small in comparison with $\sigma_0=222$ emu/g for $T=0$°K. If these departures from the theory arise from neglecting higher-order terms $\sim\sigma^6$, etc., in the free energy, then we need only consider the low-field region. In so doing we measure $\gamma_c=3.9\times 10^9 g^2cm^{-6}G^{-2}$ at $T=T_c$ and obtain $C_c=2.8\times10^{-9}$ g/G^{2}cm^3. We also obtain $A_c=12.6$ g/cm^{3}deg from magnetization data (not shown). Combining A_c and C_c in Eq. (17) provides the result $dT_c/dP=0.22$ deg/kbar. This value is much smaller than 0.34 deg/kbar derived from the magnetic Grüneisen constant obtained in I.[2] It also disagrees with results of three independent pressure experiments[16] giving the values 0.46,[16] 0.40,[17] and 0.37[18] all within ±0.1 deg/kbar.

Applying Eq. (17) to temperatures above T_c where σ is smaller should increase the validity of the termination of the power-series expansion. In particular, λ_f versus σ^2 observed at 77.5°K (Fig. 7) shows a more nearly linear behavior and gives $\gamma_c=8.2\times10^{-9}$. At this temperature A_c is unchanged and $C_c=6.0\times10^{-9}$. These combine to give $dT_c/dP=0.48$ deg/kbar. Thus we find that dT_c/dP evaluated for EuO using the Landau-Belov theory varies with temperature (see inset, Fig. 8). This result contrasts with the temperature-independent magnetic Grüneisen constant and the value $dT_c/dP=0.34$°K/kbar obtained in I by comparing data of expansivity and specific heat over a wide temperature range. Even at temperatures greater than T_c where dT_c/dP appears to level off to a constant value (inset, Fig. 8) and where the initial magnetization bebecomes small so that termination of the power series in the Landau-Belov free-energy expansion is more acceptable, the analysis gives a value $dT_c/dP=0.5$ deg/kbar which appears too large by comparison with the other experiments mentioned.

SUMMARY

From measurements of induced magnetostrain in a single-crystal disk of EuO we have obtained the linear and isotropic (volume) components of magnetostriction. Data are presented with considerable detail because the volume magnetostriction of a cubic ferromagnet reflects isotropic spin-spin correlations while the linear magnetostriction measures longitudinal spin correlations. Comparison of observations on EuO with quantum statistical theories appears particularly meaningful because EuO is a uniquely ideal Heisenberg ferromagnet with S ground-state magnetic ions, each situated on cubic sites and having predominantly nearest-neighbor exchange forces whose variation with temperature by means of thermal lattice contraction is negligible. That the volume magnetostrain is related *directly* to isotropic spin-spin correlations is implicit in the magnetic Grüneisen parameter for EuO being independent of temperature, as was shown in I.

We find here that the coefficients λ_{100} and λ_{111} of linear magnetostriction are expressible as a unique function of the observed magnetization. The function which fits the data is a 1:1 superposition of the expressions $\hat{I}_{5/2}(\mathfrak{L}^{-1}(m))$ and m^2, where $m=\sigma(H,T)/\sigma(0,0)$ is the reduced magnetization. These expressions describe longitudinal spin correlations for single-ion and two-ion interactions, respectively. It seems plausible, then, that half the strength of observed magnetoelastic coupling coefficients $b_1(0)=31\times10^6$ dyn/cm^2 and $b_2(0)=-86\times10^6$ dyn/cm^2 for $T=0$°K arises from strain-dependent single-ion interaction with the crystal field, while the other half may arise from strain-dependent two-ion energies, i.e., dipolar and pseudodipolar effects.

Although for the volume magnetostriction the theory is less exact, the character of the data of λ_f versus H_i in Fig. 2 is similar to theoretical curves computed for EuS by E. and H. B. Callen using a two-particle cluster theory. To provide a testing ground for future theoretical work, we have also presented the total isotropic magnetoelastic strain $(\delta l/l_0)_{me}$ and magnetoelastic expansivity α_{me} versus temperature for fixed internal magnetic fields. The former may be considered to describe the relative temperature variation of the internal magnetic energy in the presence of a field, while the latter reveals the manner of suppression of the λ anomaly by a magnetic field.

Finally, we have compared the forced volume magnetostriction and the magnetization, each determined as a function of H and T, with predictions of the Landau-Belov phenomenological theory describing a second-order phase transition. The comparison leads to a pressure derivative $\partial T_c/\partial p$ for the ordering temperature of EuO that changes with temperature. This variation is inconsistent with a magnetic Grüneisen constant $\partial\ln J/\partial\ln V$ being independent of temperature as determined previously in I unless lattice compressibility, which relates these quantities, also varies with temperature. Such a behavior appears unlikely for this relatively incompressible and high-melting-point material.

ACKNOWLEDGMENTS

We are particularly indebted to C. F. Guerci and M. W. Shafer for their considerable effort in growing the suitably large and good-quality single crystal of EuO.

[16] R. Stevenson and M. C. Robinson, Can. J. Phys. **43**, 1744 (1965).
[17] G. K. Sokolova, K. M. Demchuk, K. P. Rodionov, and A. A. Samakhvalov, Zh. Eksperim. i Teor. Fiz. **49**, 452 (1965) [English transl.: Soviet Phys.—JETP **22**, 317 (1966)].
[18] D. B. McWhan, D. B. Souers, and G. Jura, Phys. Rev. **143**, 385 (1966).