Anisotropic magnetostriction of $CeAl_2$ near its antiferromagnetic transition

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The magnetostriction of polycrystalline samples of $CeAl_2$ has been measured as a function of temperature and magnetic field in the vicinity of its low-temperature phase transition. We find that the magnetostrictive response is dominantly anisotropic, reflecting the anisotropy of the $4f$ charge distribution. The volume magnetostriction upon entering the low-temperature phase (LTP) precludes any significant valence change of the Ce atoms. The anisotropic magnetostriction in the LTP indicates the development of a spontaneous antiferromagnetic (AF) ordering transverse to the external field. Studies of the orientation of this transverse AF polarization show both irreproducible domain orientation in field and a reversible orientation due apparently to a continuous microscopic rotation of the AF order parameter.

I. INTRODUCTION

In a Kondo lattice (or perhaps better an Anderson lattice) one has a localized level with energy E_f in some sense close to the Fermi energy ϵ_F of a conduction band. Allowing the hybridization (or mixing) of the local and conduction electrons leads to an effective local level energy width Δ.

Within this problem it is convenient both theoretically and experimentally to draw a distinction between the local-moment regime, where $E_f - \epsilon_F > \Delta$, and the strongly mixed valent regime where $E_f - \epsilon_F \lesssim \Delta$.[1] However it must be emphasized that these are two limiting regimes of the same problem and that in general a continuous passage between these limits is possible. Examples of experimental systems in the strongly mixed-valence regime are α-Ce,[2,3] and SmS (gold phase).[4,5] In the strongly mixed-valence regime the f-level occupation number deviates from integral value by an amount on the order of 0.5 of an electron. Examples of experimental systems in the local-moment regime, often referred to as concentrated Kondo systems, are γ-Ce,[2,3] β-Ce,[3] $CeAl_3$,[6] and $CeAl_2$.[7,8] These systems tend to deviate by 0.1 of an electron or less from the normal integral valence (3^+ in the case of Ce).

For systems in the local-moment regime both ordered magnetic (e.g., $CeAl_2$)[9,10] and nonmagnetic (e.g., $CeAl_3$)[6] ground states have been found. The two opposing views of the nonmagnetic ground state for Ce in these systems are (i) that it is an array of spin-compensated Kondo singlets[11] and (ii) that the local $4f$ states are broadened into Lorentzians in the sense of the virtual bound states of Friedel and that at low temperatures these virtual bound states form a band.[6] A restatement of the latter of these two viewpoints is essentially that the ground state is mixed valent with a very narrow f-sd hybridized band[12] (weakly mixed valent).

$CeAl_2$ has been shown to have a magnetically ordered ground state.[9,10,13,14] However, many experiments have been interpreted by placing $CeAl_2$ on the borderline of a nonmagnetic ground state (see below). The experimental and theoretical ground for such borderline systems is not yet firmly established. Thorough studies are needed before conclusions regarding the presence or competition of magnetic and nonmagnetic interactions can be drawn.

This paper will deal with a detailed study of the anisotropic magnetostriction of $CeAl_2$ at low temperatures. In Sec. II we give a review of results on the $CeAl_2$ system. In Sec. III we discuss the anisotropic magnetoelastic coupling typical of rare-earth systems and theoretical predictions for polycrystalline magnetostriction. In Sec. IV we discuss sample preparation and experimental procedure. In Sec. V the magnetostrictive response of $CeAl_2$ in its paramagnetic and in its antiferromagnetic low-temperature phase (LTP) is presented and discussed. The transverse and longitudinal thermal expansion are compared in Sec. VI. In Sec. VII our conclusions are summarized briefly and some important open questions are mentioned.

II. INTRODUCTION TO $CeAl_2$

By comparing the lattice constant of $CeAl_2$ to the other rare earth dialuminides one obtains a valence estimate of about 3.1^+ for Ce.[7] This small deviation from 3^+ valence is barely beyond the intrinsic uncertainties of such a lattice parameter valence estimate. The classification of $CeAl_2$ in the local-moment regime (in the sense discusssed above) is consistent with the lattice-parameter measurement. From the thermal expansion measurements of Walker *et al.*[8] and those

of the authors it is also clear that $CeAl_2$ remains in the local-moment regime down to at least 1.3 °K.

The cubic crystalline electric field (CEF) in $CeAl_2$ splits the $J = \frac{5}{2}$ level of Ce into a Γ_7 ground-state doublet lying about 100 °K below the excited Γ_8 quartet.[15] The poor quantitative fits of the specific heat[16] and susceptibility[17] for this CEF level structure have been attributed to partial conduction electron compensation of the local moments (the Kondo effect). Cornut and Coqblin have fit the resistivity of $CeAl_2$ between 10 and 300 °K to a CEF modified Kondo effect.[18] It should be noted that Cornut and Coqblin attributed the resistivity rise below 13 °K to critical scattering above a magnetic transition. Buschow and van Daal believed that same structure to be due to the Kondo effect of the Γ_7 ground state.[19]

The most definitive published work on the CEF structure of $CeAl_2$ is the inelastic neutron scattering work of Lowenhaupt and Steglich.[15] They observed Γ_7 and Γ_8 levels with finite widths separated by 9 meV. They interpreted the internal widths of these levels in terms of the Kondo effect only and obtained a Kondo temperature for the upper level of 25 °K and a Kondo temperature for the Γ_7 ground state of "the order of degrees Kelvin."[20] They further saw the amplitude of the quasielastic scattering within the Γ_8 quartet disappear at low temperature as the Γ_8 level depopulated.

The low-temperature properties of $CeAl_2$ are dominated by a set of phase transition anomalies below 5 °K and 60 kOe. The experimental situation at the onset of our work is well described by Walker *et al.*[8] The relation of the various low-temperature anomalies was sufficiently unclear for Walker *et al.*[8] to conclude that two separate phase transitions were involved and for others to question whether any phase transition at all occurred.[11] Although the presence of magnetic order in the low-temperature phase (LTP) of $CeAl_2$ had been inferred by Walker *et al.*[8] and by Croft, Zoric, Markovics, and Parks[13] the reported absence of magnetic Bragg reflections[21] in the LTP left the ordering question open. Only very recently has ordering been confirmed by Benoit *et al.*[9] through anisotropic γ-ray emission of ^{137}Ce in the LTP and by Barbara *et al.*[10] through neutron scattering experiments, the latter study revealing a "modulated antiferromagnetic structure."

III. RARE-EARTH MAGNETOSTRICTION: POLYCRYSTALLINE MAGNETOSTRICTION

Much of the information about the low-temperature properties of $CeAl_2$ presented in this paper are based upon magnetostriction and finite-field thermal-expansion measurements. At the onset it should be emphasized that the magnetostrictive coupling in $CeAl_2$ is large, typical of rare-earth atoms in the solid state and is rooted in the strong anisotropy of the $4f$ wave function. A strong spin-orbit interaction makes the total angular momentum $J = L + S$ the good quantum number for the rare earths and casts the anisotropic character of the $4f$ orbital into the local magnetic moment.

Cullen and Clark[22] have recently given a detailed and physically tangible treatment of the anisotropic magnetostriction in the RFe_2 (R is a rare earth) series, the latter being isostructural with the RAl_2 series; although it should be noted that the details of the distortive response need not be the same in these two systems, Barbara *et al.*[23] have shown spontaneous distortions of the order of $\Delta l/l \sim 10^{-3}$ at the Curie points of $TbAl_2$, $NdAl_2$, and $DyAl_2$. For the latter two of these compounds the easy axis of magnetization is [100] and the distortion is tetragonal with the conjugate elastic modulus being $C_{11} - C_{12}$.[23] $TbAl_2$, on the other hand, is a [111] easy axis and the distortion is rhombohedral with the conjugate elastic modulus being C_{44}.[23] In the case of Tb the charge cloud in the polarized state is "pancake-shaped (oblate)"[22] and transverse to the direction of the moment. The distortion of $TbAl_2$ (and $TbFe_2$) is such that along the direction of spontaneous magnetization a contraction is observed. The charge distribution of the polarized Γ_7 CEF ground state of $CeAl_2$, shown in Fig. 1, shows a charge distribution weighted

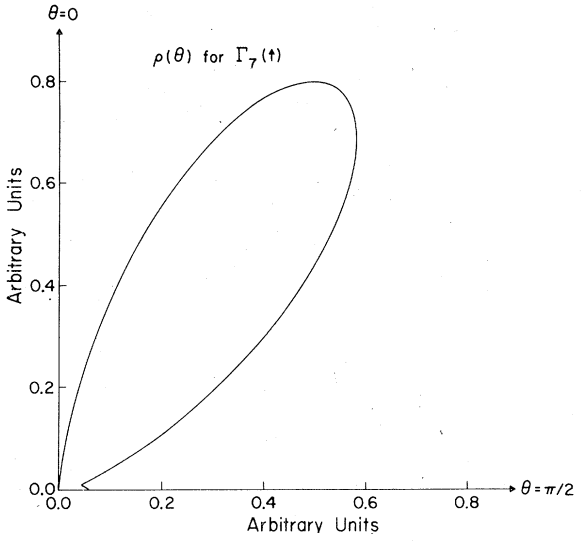

FIG. 1. Polar plot of charge distribution as a function of azimuthal angle for the fully polarized Γ_7 state of $CeAl_2$ with the magnetic moment along the vertical axis ($\theta = 0$).

toward the direction of the magnetic moment (z direction). The direction of the large lobe of the charge distribution is about 34.5° from the z axis. The ratio of the projection on the z ($\theta=0$) direction to the projection on the $\hat{r}$ ($\theta=\frac{1}{2}\pi$) direction is 1.37. Thus, the charge distribution asymmetry is opposite to that of Tb in $TbAl_2$ and of much smaller magnitude. The field-induced distortion in $CeAl_2$, as we shall see, is such that an elongation is seen along the direction of the local moment, as might be expected from the $TbAl_2$ analogy.

The most general theoretical treatment of magnetostriction is that of Callen and Callen[24] (CC). Their treatment is based upon a free energy which includes an elastic energy (of second degree in the strains), an isotropic spin-spin ferromagnetic exchange energy (of second degree in separate site spin variables), a spin anisotropy energy (of second and fourth degree in spin variables), and a magnetoelastic coupling energy (of zero and second degree in spin variables and linear in the strains). In the CC theory for the case of a cubic system with four independent magnetoelastic constants the relative change in length, of a single crystal (measured along a direction $\hat{\beta}$ with the magnetic field along the $\hat{\alpha}$ direction), can be written

$$\frac{\Delta l}{l}(\hat{\alpha}\hat{\beta}) = \frac{\lambda^{\alpha}-\lambda_1^{\gamma}}{3} + (2\lambda^{\epsilon}-\lambda_1^{\gamma})\sum_i \alpha_i^2\beta_i^2 + 2\lambda^{\epsilon}\sum_{i\neq j}\alpha_i\beta_i\alpha_j\beta_j + \tfrac{3}{2}\lambda_2^{\gamma}[(\alpha_1^2-\alpha_y^2)(\beta_z-\tfrac{1}{3}) - (\alpha_z^2-\tfrac{1}{3})(\beta_x^2-\beta_y^2)]. \quad (1)$$

We have written this expression in this form, rather than that given by CC, to enable the reader to compare the results to the simpler but less general theory of Averbach and Segransan[25] (AS). Comparison with the three magnetoelastic constant theory of AS allows the identification of λ^{α} with the homogeneous (volume) deformation, λ^{ϵ} with the distortion of trigonal symmetry and λ_1^{γ} with a tetragonal response. (In the case of three independent magnetoelastic coupling constants the subscript in λ_1^{γ} can be dropped.) At this juncture it should be emphasized that the C-15 structure of $CeAl_2$ has high enough symmetry to support only three magnetostrictive constants. The more general discussion given here is included because the authors have not seen it previously in print and because there may be interesting systems of lower symmetry which cannot be easily prepared in single-crystal form.

If one assumes a spherical distribution of crystallites in a polycrystalline sample the polycrystalline expression for $\Delta l(\gamma)/l$ is given by

$$\frac{\Delta l}{l}(\gamma) = \tfrac{1}{3}\lambda^{\alpha} + \tfrac{2}{5}(\tfrac{1}{3}\lambda_1^{\gamma}+\lambda^{\epsilon})(-1+3\cos^2\gamma), \quad (2)$$

where γ is the angle between $\hat{\alpha}$ and $\hat{\beta}$. There are two points we would like to make about this result. First, the fourth magnetostrictive response λ_2^{γ} drops out in the spherical average and therefore one cannot tell from a polycrystalline measurement the difference between cubic groups which support three or four magnetoelastic constants. Second, the polycrystalline magnetostriction has only two independent variables, the homogeneous strain $\frac{1}{3}\lambda^{\alpha}$ and the distortive strain $\frac{2}{5}(\lambda_1^{\gamma}\frac{1}{3}+\lambda^{\epsilon})$ (a weighted sum of trigonal and tetragonal strain). These two independent quantities are conveniently expressed in terms of the longitudinal $\Delta l/l$ [i.e., $\Delta l/l = \Delta l/l$ ($\gamma=0$)] and transverse $\Delta r/r$ [i.e., $\Delta r/r = \Delta l/l$ ($\gamma=\frac{1}{2}\pi$)] magnetostriction;

$$\frac{\Delta v}{v} = \frac{\Delta l}{l} + 2\,\frac{\Delta r}{r} = \lambda^{\alpha}, \quad (3a)$$

$$\frac{\Delta \xi}{\xi} = \frac{\Delta l}{l} - \frac{\Delta r}{r} = \frac{4}{5}\left(\frac{\lambda_1^{\gamma}}{3}+\lambda^{\epsilon}\right), \quad (3b)$$

where $\Delta v/v$ is the homogeneous volume magnetostriction and $\Delta\xi/\xi$ is a measure of the distortive magnetostriction.

The field and temperature dependence of the magnetostrictive responses (i.e., the λ's) in the CC theory is in general complicated and contains two terms—one due to the single-ion local-anisotropy effects and one due to two ion-correlation effects. The functional dependence of the single-ion term is that of the ratio of two Bessel functions whose arguments are $\mu H_i/kT$, where μ is the local magnetic moment, H_i is the internal field, k is Boltzmann's constant, and T is the temperature. The two ion-term couples to the two ion-correlation function, which in mean-field approximation reduces to the magnetization squared.

IV. SAMPLE PREPARATION AND EXPERIMENTAL TECHNIQUES

Polycrystalline $CeAl_2$ smaples were prepared by arc melting stoichometric amounts of 0.9999 pure Ce and 0.99999 pure Al in an argon arc furnace with a 10-psi overpressure of Ar. The samples were subsequently wrapped in tantalum foil, sealed in evacuated quartz tubes along with Zr getters and annealed for two weeks at 770 °C. The samples were in approximately 1-cm boules and contained individual crystallites of the order of 0.1 mm. X-ray analysis showed the samples to exhibit only the $MgCu_2$ structure with lattice constant 8.054.[26]

All the thermal expansion and magnetostriction measurements were made with the standard capa-

citive dilatometry technique.[27] These length measurements were made relative to the fused quartz from which the capacitance cell we constructed. The measurements themselves were made with a General Radio 1615A capacitance bridge driven by and monitored by 124 PAR lock-in amplifier. All temperature measurements were made with a field-insensitive calibrated carbon glass (CG) thermometer obtained from Lake Shore Cryotronics, Inc. The magnetoresistance shift of the CG thermometer in 115 kOe and near 4 °K corresponded to a temperature shift of less than 0.25 °K and was neglected. To insure temperature homogeneity all measurements were made in ^{4}He exchange gas.

Magnetic fields below 54 kOe were achieved with a conventional medium field superconducting solenoid. The fields in this solenoid were determined by measuring the current supplied to the solenoid and using the results of an independent calibration. All measurements were made in constant or increasing field to eliminate trapped flux ambiguities. Measurements in fields up to 115 kOe were made in a superconducting solenoid custom manufactured by the Intermagnetics Co. The magnetoresistance of copper was used to measure the field in this magnet. An electromagnet which could be rotated through 180° to a precision of 0.1° and with field control precise to 1 Oe was used in the studies of low-field hysteresis and angular dependence of the magnetorestriction.[28]

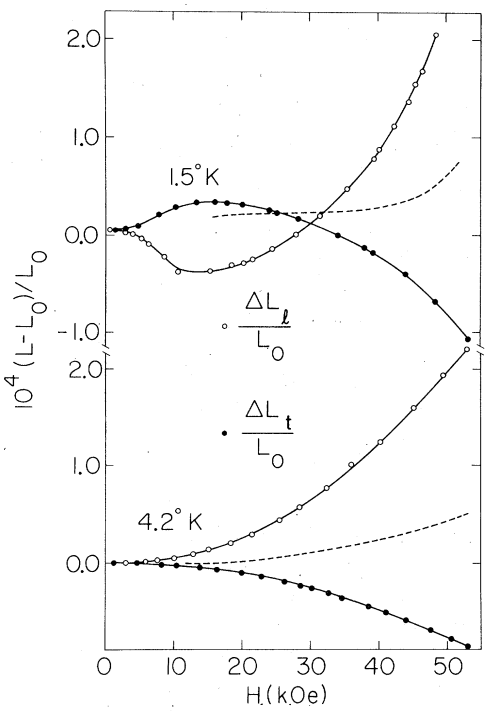

FIG. 2. Longitudinal magnetostriction (change of length parallel to field) (open circles) and transverse magnetostriction (closed circles) in the LTP (1.5 °K) and above the LTP (4.2 °K). Dotted lines: estimates of volume magnetostriction using Eq. (3a). L_0 is sample length at 4.2 °K and $H=0$ in this and subsequent figures.

V. MAGNETOSTRICTION IN $CeAl_2$

In Fig. 2 we show the length measured longitudinal and transverse to the magnetic field at temperatures $T=1.5$ °K and $T=4.2$ °K for a polycrystalline sample of $CeAl_2$. At 4.2 °K one is above the transition into the LTP and at 1.5 °K one is in the LTP. There are five points to note in this figure: (i) Above the LTP (at 4.2 °K) the longitudinal and transverse magnetostriction coefficients [the slopes of the $L(H)$ curves] have opposite sign with an expansion occurring longitudinal to the field. This is consistent with the domination of the distortive magnetoelastic coupling in Eq. (2) (ii) At 4.2 °K the relative voume ($\Delta v/v$) as computed from Eq. (3a), shown as a dotted line in the lower portion of Fig. 2, increases in increasing field. The total change in $\Delta v/v$ is small and of the order of 0.5×10^{-4} at 50 kOe. (iii) In the low temperature phase, at $T=1.5$ °K, the signs of the transverse and longitudinal magnetostriction in fields less than 15 kOe reverse. The magnitude of the distortion at 15 kOe is of the order of 0.5×10^{-4}, which is much smaller than in the ferromagnetic RAl_2 compounds (R = rare earth). (iv) The magnetostriction shows a distinct nonlinearity in fields below 10 kOe. This low field nonlinearity is related to the tendency for a spontaneous antiferromagnetic (AF) polarization to align transverse to an external magnetic field. (This tendency is discussed in Sec. VI in regard to the finite-field thermal expansion.) This alignment can occur either by microscopic changes in the order-parameter orientation or by the forced rotation of domains. (v) If one computes the volume change in the LTP via Eq. (3a), the volume again increases in increasing field (dashed line at $T=1.5$ °K). In fields below 15 kOe, when the domains are not uniformly oriented with respect to the external field, the estimation of the volume via Eq. (3a) is certainly quantitatively incorrect. The volume estimate above 15 kOe is, however, probably qualitatively correct. The volume estimate in the LTP shows a weak field dependence below 40 kOe. As one approaches the phase boundary in high field (about 56 kOe) the volume shows a rather rapid rise. The LTP therefore appears to have a smaller volume than the polarized paramagnetic state.

The longitudinal magnetostriction at 4.2 and 1.5 °K in fields up to 115 kOe is shown in Fig. 3.

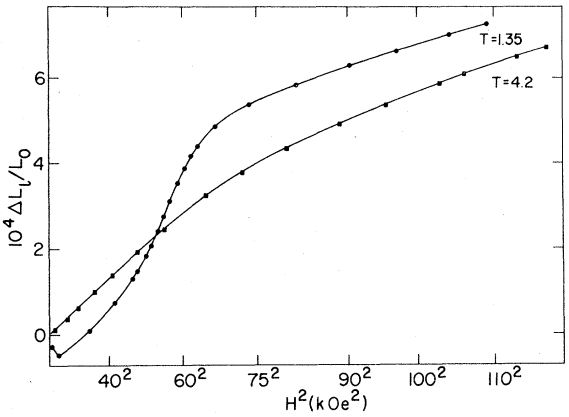

FIG. 3. Longitudinal magnetostriction versus square of magnetic field at 1.35 and 4.2 °K.

There are three points we would like to make regarding these curves. (a) At high field the same lack of saturation as seen in the magnetization measurements of Barabara *et al.*[32] is evident. (b) The high-field phase boundary presumably occurs where the slope of the ΔL vs H curve is maximum, around 56 kOe. (The precise critical field and strength of the nonlinearity in magnetization have been shown[32] for a single crystal to be direction dependent.) (c) It is important to note that although the $T = 1.5$ °K sweep was made by decreasing the field from 115 kOe the upturn in the length below 15 kOe still appears. This would indicate that the transverse reorientation of the AF polarization is not simply domain reorientation since this would be irreversible.

The details of the reproducible and irreproducible parts of the low-field magnetostriction in $CeAl_2$ are shown in Fig. 4. Curve 4(a) shows the totally reversible longitudinal magnetostriction at 4.2 °K (i.e., above the LTP). The sample was cooled in zero field to point *A* in Fig. 4(b) and the field turned slowly up to 9.5 kOe, to point *B*. The field was then lowered slowly tracing out the line *B-C*. The fact that the length at point *C* is different than that at point *A* is due presumably to irreversible domian orientation. The field was then turned up to −9.5 kOe, to point *D*, and back to zero field yielding line *D-E*. The zero field points *C* and *E* now reproduce. Further cycling to ±9.5 kOe essentially reproduces the *C-D-E* hysteresis loop (or its mirror image). Thus the initial domain reorientation along curve *A-B* was always maintained. (This was to be expected since the same transverse orientation occurs for either sign of the external field.)

Another dramatic illustration of the irreversible domain reorientation can be seen in the transverse magnetostriction. The field cycles *A-B-C-D-E* were made with the field oriented longitudinal (in the direction $\hat{l}$ for clarity) to the length measurements. After these longitudinal cycles a field was applied transverse (in the $\hat{r}$ direction) to the $\hat{l}$ direction. The transverse magnetostriction starting from zero field is shown in Fig. 4(c). Since the original irreversible domain orientation was made with a preferred direction $\hat{l}$, the transverse curve *F-G-H* again shows irreversible domain reorientation hysteresis. The same reproducible hysteresis is also seen in loop *H-I-J*.

It should be noted that this magnetostrictive sensing of domain reorientation is a very sensitive macroscopic probe of the ordered nature of the LTP. The attempt by Cornut *et al.*[7] to observe remnant magnetization due to imprefections in the AF state was not successful. In the work of Benoit *et al.*[9] no γ-ray anisotropy (i.e., domain orientation) could be sensed with less than 34 kOe, whereas the irreversible domain orientation is clear in our work below 10 kOe. That there is more going on than simple domain orientation

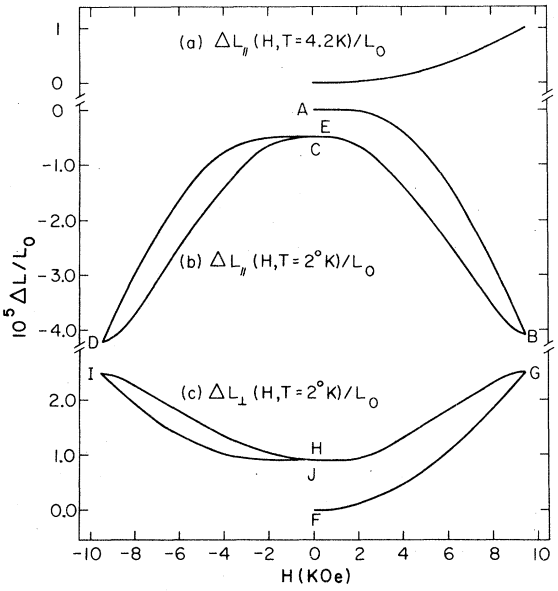

FIG. 4. (a) Totally reversible longitudinal magnetostriction at 4.2 °K. (b) Longitudinal magnetostriction at 2.0 °K (in the LTP). Point A was reached by cooling in zero field. The field was then turned up to +9.5 kOe (point B), then down to zero field (point C), then up to −9.5 kOe (point D), then down to zero field (point E). The irreversible domain orientation in cycle A-B-C is evident. The reversible hysteresis loop C-D-E, due presumably to domain edge effects, is also exhibited. (c) Transverse magnetostriction at $T = 2$ °K. These measurements immediately followed those in (b). The curve F-G-H shows the irreversible domain reorientation in the A-B-C cycle. Again, subsequent cycles show the reproducible type loop H-I-J.

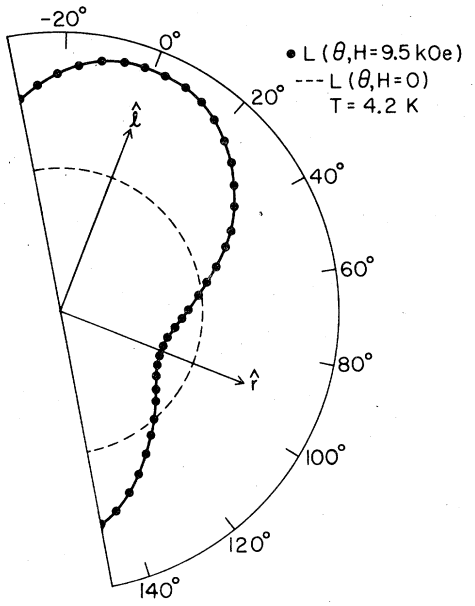

FIG. 5. Polar plot of magnetostriction (solid points) in arbitrary units versus angle of length measurement relative to applied field. The $\hat{l}$ (longitudinal) and $\hat{r}$ directions are indicated. The solid line is a fit to the empirical form $A+B\cos^2\gamma$. The dotted line is the length in zero field.

in low field in the LTP is indicated by the negative magnetostriction occurring both when lowering field from very high field (previously noted in Fig. 3) and when cycling around the reproducible C-D-E low field hysteresis loop [Fig. 4(a)]. The major part of the transverse reorientation of the AF polarization is reproducible and from this we presume it is microscopic in origin.

The microscopic order parameter reorientation could be first order, like in a spin-flop transition or it could be continuous. The reproducible low-field hysteresis could be due to a domain average of first-order reorientation transitions with strongly anisotropic critical fields. On the other hand, it could be due to domain-boundary effects with the bulk reorientation occurring continuously. Magnetostriction measurements on single crystals by our group[29] and by the Koln[30] group show no manifestly first-order jumps. Further, both the domain-orientation effect and reproducible hysteresis loops are also seen in single-crystal measurements.[29] These last two facts simply emphasize that even a single crystal is of polydomain character in the LTP in low enough field. What appears to be going on is a continuous microscopic reorientation of the AF polarization in low field with reproducible hysteresis being caused by domain-boundary effects.

As mentioned previously the magnetostriction above the LTP is totally reproducible both as a function of field cycling and field orientation. Fig. 5 shows a polar plot of the magnetostriction at 4.2 °K as a function of the angle between the field direction and the direction along which the magnetostriction is being measured. The longitudinal ($\hat{l}$) and transverse ($\hat{r}$) directions are at the maximum and minimum of the magnetostriction respectively. The angular dependence obeys the functional form $A+B\cos^2\gamma$ with $A=-40$ and $B=180$ in arbitrary units.

Comparing the empirical fit to the theoretical form [(Eq. (2)], one finds that at $T=4.2$ °K and $H=9.5$ kOe the strain variables have the values, $\lambda^\alpha=60$ and $\frac{4}{5}(\frac{1}{3}\lambda_1^\gamma+\lambda^\epsilon)=120$ in arbitrary units. As expected the distortive strain is much larger than the homogeneous volume strain. As mentioned earlier, from the form of Eq. (2) it is clear that polycrystalline magnetostriction measurements can determine only the homogeneous and total distortive strain but cannot differentiate between trigonal (λ^ϵ) and tetrahedral (λ^γ) strain. The [111] polarization direction of the ordering in zero field in the LTP[10] would lead one to expect a spontaneous trigonal (rhombohedral) distortion upon entering the LTP in zero field. The critical softening observed by Godet and Purwins[31] in c_{44} is in agreement with this expectation, since this is the appropriate modulus conjugate to a trigonal distortion.

VI. TRANSVERSE COMPARED TO LONGITUDINAL THERMAL EXPANSION

The results of measurements of length transverse and longitudinal to the field versus temperature in fields of 15, 48, and 53 kOe are shown in Fig. 6. In Fig. 7, estimates, using Eq. (3a), Eq. (3b) and the curves in Fig. 6, of both the distortive and homogeneous contributions to the magnetostriction are shown. The scales for $\Delta v/v$ and $\Delta\epsilon/\epsilon$ are the same and the results for $\Delta v/v$ is inset between the $\Delta\epsilon/\epsilon$ plots so that the separations between the $\Delta\epsilon/\epsilon$ curves at different fields are quantitatively correct.

The distortive magnetostriction is obviously strongly temperature dependent. Above the LTP, $\Delta\epsilon/\epsilon$ is large and positive with an expansion along the direction of the field which increases in increasing field and decreasing temperature. At 15 kOe, when one enters the LTP, $\Delta\epsilon/\epsilon$ changes sign, i.e., longitudinal to the field there is a contraction. The simplest explanation for this sign reversal in $\Delta\epsilon/\epsilon$ is that a spontaneous local magnetic polarization transverse to the external field develops at the transition.

In an antiferromagnet when an external field is

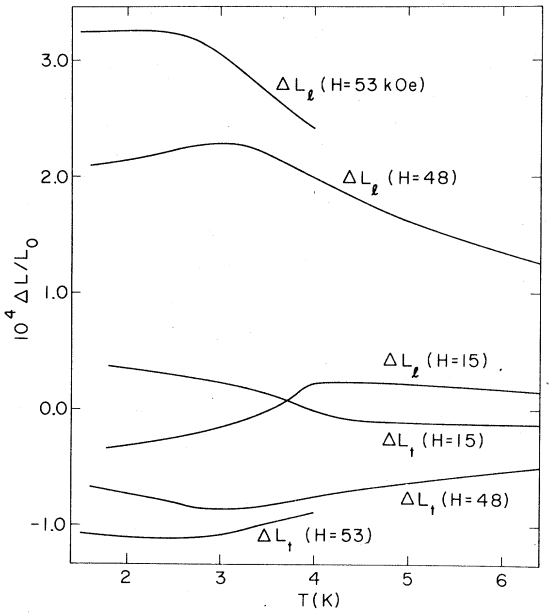

FIG. 6. Longitudinal (ΔL_l) and transverse (ΔL_t) magnetostriction versus temperature for different field values.

along the direction of polarization of the AF order parameter the magnetic response is the energetically costly one of reversing some spins, in direct competition with the ordering. On the other hand a field transverse to the AF polarization can simply cant the spins—hence, the much larger transverse susceptibility and preferred transverse ordering for an AF in an applied field. At 48 and 53 kOe the distortion $\Delta\epsilon/\epsilon$ is always positive but the development of a transverse component to $\langle\vec{S}_i\rangle$ is apparent from the downturn in the $\Delta\epsilon/\epsilon$ curves at the transition into the LTP.

In Fig. 7 (inset) the general tendency for the volume to increase upon lowering temperature even upon entering the LTP is evident. Again the use of Eq. (3) for estimating the volume in the LTP must be done with reservations. The volume expansion is very small, compared to the scale of volume changes corresponding to valence changes. The volume expansions observed, if attributed to a change in f-electron occupation would correspond to an increase of only about 0.0002 and 0.0013 of an electron in zero and 53 kOe, respectively, which is too small to be regarded as significant.

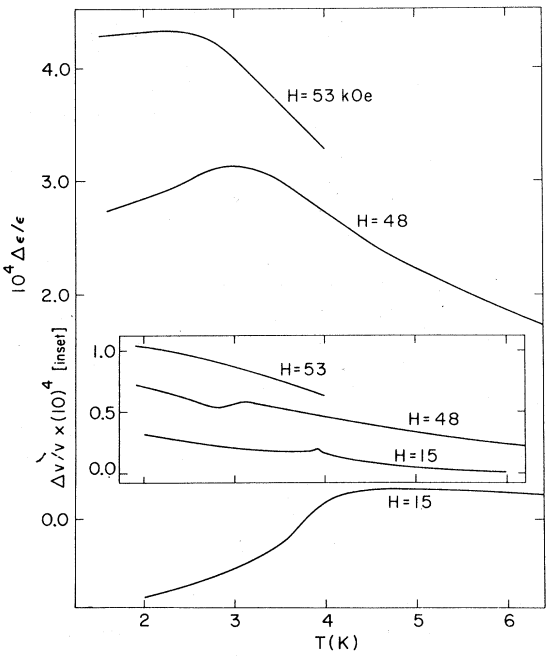

FIG. 7. Distortive component of the strain $\Delta\epsilon/\epsilon$ vs temperature. Inset: homogeneous volume strain $\Delta v/v$ vs temperature.

VII. CONCLUSION

We have shown that the magnetostriction of $CeAl_2$ in the paramagnetic phase is predominantly distortive in character which is typical of rare-earth local-moment systems. In the low temperature phase (LTP) of $CeAl_2$ the magnetostriction is indicative of antiferromagnetic ordering and is dominated by the development of a spontaneous AF polarization transverse to the external field. Part of the magnetostriction in the LTP was seen to be due to irreversible domain reorientation. However, the largest part of the magnetostriction associated with the transverse reorientation of the AF polarization is reversible. This presents the possibility that the reorientation is microscopic and continuous. This would be in contrast to the TmSe system where the reorientation is a discontinuous and totally irreversible (along the principal axis) domain reorientation.[33,34] In the TmSe system these properties have been attributed to strong anisotropy.[34] In $CeAl_2$ we may have the opposite case of weak anisotropy. Indeed the saturation magnetization along the body diagonal and face diagonal in $CeAl_2$ are quite close. The rotational of the ordering from the easy [111] axis to a face diagonal axis perpendicular to [111] should therefore cost very little anisotropy energy.

The ordered character of the $CeAl_2$ ground state is now well established. There are, however, a number of interesting questions which remain open for this experimental system. From our work there is the question of the basic nature of the AF polarization reorientation process.

Elastic neutron scattering experiments in fields between 0 and 15 kOe should provide the decisive answer here. More interesting to some is the question of how the tendency toward Kondo singlet or toward virtual bound-state formation competes with the magnetic-exchange interaction. Whether this competition is responsible for the instability of ferromagnetism in $CeAl_2$ (the other trivalent rare earth dialumindes are ferromagnetic) or causes the modulated moment structure seen by Barbara *et al.*[10] remains to be seen.

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