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Magnetostriction

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This paper reviews work since 1961 on magnetostriction, including both its sources and its temperature and magnetic field dependence beyond saturation (forced magnetostriction). A statistical theory of the magnetostriction of ferromagnets is in fine agreement with experiments on nickel and on heavy rare-earth metals, which are reviewed in detail. A different mechanism or two has been invoked to describe the behavior of iron. The paper sketches the origins of magnetoelastic coupling, particularly in rare-earth metals, and describes a phenomenological spin Hamiltonian approach to nonmetals. The theory is then extended to ferrimagnets, whose magnetostrictive behavior is described. Experiments on spinels and garnets are reviewed. The meager literature of the magnetostriction of antiferromagnets is surveyed, and the review ends with a brief description of oscillatory magnetostriction in diamagnets.

1. INTRODUCTION

Magnetostriction is the lattice deformation which accompanies magnetization. This paper reviews some work on magnetostriction since 1961. It is far from exhaustive; polycrystal measurements, domain, dynamic magnetoelastic, and size effects are excluded. Our concern is static, saturation, spatially uniform magnetostriction of single crystals.

Designating $\boldsymbol{\alpha}$ as the direction of magnetization (which we consider to lie along an external magnetic field) with respect to the crystal axes, the fractional change in length of a cubic crystal in direction $\boldsymbol{\beta}$ is given by simple symmetry considerations to be of the form[1]

$$\delta l/l=\lambda_0(T,H)+\tfrac{3}{2}\lambda_{100}(T,H)[\alpha_1^2\beta_1^2+\alpha_2^2\beta_2^2+\alpha_3^3\beta_3^3-\tfrac{1}{3}]$$
$$+3\lambda_{111}(T,H)(\alpha_1\alpha_2\beta_1\beta_2+\alpha_2\alpha_3\beta_2\beta_3+\alpha_3\alpha_1\beta_3\beta_1]+\cdots. \quad (1)$$

Similarly, a convenient form for a hexagonal crystal is

$$\delta l/l=\lambda_1^{\alpha,0}(T,H)(1-\beta_z^2)+\lambda_2^{\alpha,0}(T,H)\beta_z^2$$
$$+\lambda_1^{\alpha,2}(T,H)(\alpha_z^2-\tfrac{1}{3})(1-\beta_z^2)+\lambda_2^{\alpha,2}(T,H)(\alpha_z^2-\tfrac{1}{3})\beta_z^2$$
$$+\lambda^{\gamma,2}(T,H)\{\tfrac{1}{2}(\alpha_x^2-\alpha_y^2)(\beta_x^2-\beta_y^2)+2\alpha_x\alpha_y\beta_x\beta_y\}$$
$$+2\lambda^{\epsilon,2}(T,H)(\alpha_x\beta_x+\alpha_y\beta_y)\alpha_z\beta_z+\cdots. \quad (2)$$

Only terms of zero and second degree ($l=0, 2$) in the direction cosines of the magnetization have been included here. Our task is the determination of the magnitude, temperature and field dependence of the various λ's.

Dipolar interactions turn out to be relatively unimportant in cubic crystals, and in crystals of lower symmetry which contain degenerate ground-site ions such as cobalt and rare earths. The relevant Hamiltonian is then

$$\mathcal{H}=\mathcal{H}_{\text{exchange}}+\mathcal{H}_{\text{xtal field}}+\mathcal{H}_{\text{spin-orbit}}+\mathcal{H}_{\text{elastic}}. \quad (3)$$

Keeping terms up to second degree in spin operators, this can be transformed, when there are localized spins, to an effective spin Hamiltonian containing two-ion exchange and single-ion anisotropy terms:

$$\mathcal{H}\sim\mathbf{S}_i\cdot\mathbf{J}_{ij}\cdot\mathbf{S}_j+\mathbf{S}_i\cdot\mathbf{D}\cdot\mathbf{S}_i+\mathcal{H}_{\text{elast}}. \quad (4)$$

The second-order anisotropy term involving $\mathbf{D}$ can easily be generalized to orbitrary order l, and we shall do so when required. The coefficients $\mathbf{J}$ and $\mathbf{D}$ can change when the crystal is strained. To first order in the various strains ϵ_μ (see Ref. 1 for details),

$$\mathcal{H}\sim\mathbf{S}_i\cdot\mathbf{J}_{ij}^0\cdot\mathbf{S}_j+\mathbf{S}_i\cdot\mathbf{D}^0\cdot\mathbf{S}_i+\epsilon_\mu\mathbf{S}_i\cdot(\partial\mathbf{J}_{ij}/\partial\epsilon_\mu)\cdot\mathbf{S}_j$$
$$+\epsilon_\mu\mathbf{S}_i\cdot(\partial\mathbf{D}/\partial\epsilon_\mu)\cdot\mathbf{S}_i+\tfrac{1}{2}c_\mu\epsilon_\mu^2. \quad (5)$$

2. ISOTROPIC EXCHANGE

There are numerous possibilities. One important case is that $\mathbf{J}_{ij}^0$ is isotropic and large compared to $\mathbf{D}^0$. The two-ion term $\partial\mathbf{J}/\partial\epsilon$ can have both isotropic and anisotropic parts but only the anisotropic part of the single-ion $\partial\mathbf{D}/\partial\epsilon$ is physically significant. This viewpoint appears to describe at least nickel metal (despite the fact that it is based on a localized spin Hamiltonian), heavy rare-earth metals, and Fe^{3+} in spinels and garnets. The derivatives, $B^\mu\equiv-\partial\mathbf{D}/\partial\epsilon_\mu$ and $D^\mu\equiv-\partial\mathbf{J}/\partial\epsilon_\mu$ are called magnetoelastic constants. Treating the linear strain terms as perturbations, the first-order free energy is minimized, and

$$\epsilon_\mu\sim\delta_{\mu,\alpha}/c_\alpha\sum_{<ij>}D^\alpha(i,j)\langle\mathbf{S}_i\cdot\mathbf{S}_j\rangle_0$$
$$+\alpha_\mu/c_\mu\sum_{<ij>}\sum_l D_l^\mu(i,j)\langle Y_l^0(\mathbf{S}_i,\mathbf{S}_j)\rangle_0$$
$$+\alpha_\mu/c_\mu\sum_i\sum_l B_l^\mu\langle Y_l^0(\mathbf{S}_i)\rangle_0. \quad (6)$$

The isotropic correlation function $\langle\mathbf{S}_i\cdot\mathbf{S}_j\rangle_0$, and the two-ion and one-ion longitudinal correlation functions, $\langle Y_l^0(\mathbf{S}_i,\mathbf{S}_j)\rangle_0$ and $\langle Y_l^0(\mathbf{S}_i)\rangle_0$, are all discussed in more detail in Ref. 1. The first term is clearly related to the change in magnetic internal energy. In a cubic crystal with isotropic exchange, only volume strain ($\epsilon_\mu=\delta V/V$) induces a change in J. In a nonconductor, and when there is only a single (nearest-neighbor) exchange constant, the magnetic contribution to the thermal

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

expansion coefficient,

$$\alpha_m(T, H) = 3[\partial\lambda_0(T, H)/\partial T], \tag{7}$$

should be proportional to the magnetic part of the specific heat. This was predicted[1] for EuO and has been so observed.[2]

Although λ_0 is often about 100 times larger than λ_{100}, it is not so easily observed, because it is independent of both α and β. On the other hand, it dominates the field dependence of strains of lower symmetry. If we write, for ferro- and ferrimagnets,

$$\lambda_i(T, H) = \lambda_i(T, 0) + \lambda'(T)H, \tag{8}$$

the "forced" volume magnetostriction of a cubic crystal $\lambda_0'(T)$, because of its exchange origin, usually dominates other forced (i.e., field dependent beyond saturation) terms. Perhaps for this reason forced volume magnetostriction is often referred to simply, but ambiguously, as "forced magnetostriction," although in fact all coefficients have a forced component.

In iron-group metals the theory is of course more complicated than Eq. (6) is contrived to handle, but the forced volume magnetostriction is still of exchange origin. Stoelinga *et al.*[3] have measured the concentration and temperature dependence of λ_0' in binary FeCo and FeNi alloys. This interesting work relates λ_0', magnetization and electronic specific heat to the densities of states at the Fermi level of the exchange split spin-up and spin-down $3d$ bands. Fawcett and White[4] have measured forced $\delta l/l$ in several directions on Ni and Co single crystals at 4.2°K, but do not resolve individual magnetostrictive components. Cobalt, being uniaxial, has two fully-symmetric strains, both of which can arise from isotropic exchange (see Ref. 1). This also applies to the heavy rare-earth metals. Coleman and Pavlovic[5] have measured all six $l=0$, 2 forced components in Gd.

The second and third terms of Eq. (6) can produce nonsymmetric strains, λ_{100} and λ_{111}, in cubic crystals [Eq. (1)] and λ^{γ},[2] and λ^{ϵ},[2] [Eq. (2)] in uniaxial ones. Research has advanced on two fronts: evaluation of the temperature dependence in the statistical averages $\langle Y_l^0(\mathbf{S}_i)\rangle_0$ and $\langle Y_l^0(\mathbf{S}_i, \mathbf{S}_j)\rangle_0$, and evaluation of the temperature-independent B_l^{μ} and D_l^{μ} by atomic calculation. One can also conceive of, and correct for intrinsic temperature dependences in the magnetoelastic coefficients themselves, such as results from thermal expansion, but experience and pressure measurements have often shown these corrections to be rather small. (But see discussion of nickel in Sec. 3.)

We next discuss the temperature dependence of anisotropic magnetostriction (λ_{100}, λ_{111}, λ^{γ}, λ^{ϵ}), anisotropic in that it depends upon the direction of magnetization. Evaluation of magnetoelastic coefficients is discussed later.

3. TEMPERATURE DEPENDENCE, FERROMAGNETS

Recall that we have temporarily assumed an isotropic exchange interation, such as $J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j$, and let us consider perturbative strain terms vaguely of the form $-B_l^{\mu}Y_l^m(\mathbf{S}_i)\epsilon_{\mu}$ (for more detail see Ref. 1). Equation (6) then follows, and $\lambda^l(T, H)$ is proportional to $\langle Y_l^0(\mathbf{S}_i)\rangle_0$. Kittel and Van Vleck[6] pointed out that this same quantity occurs in the temperature dependence of magnetic anisotropy. At sufficiently low temperatures,

$$\langle Y_l^0(\mathbf{S}_i)\rangle_T/\langle Y_l^0(\mathbf{S}_i)\rangle_0 \cong [M(T)/M(0)]^{l(l+1)/2} \equiv m(T)^{l(l+1)/2}; \qquad m(T)\cong 1. \tag{9}$$

This famous $l(l+1)/2$ power law[6] has been derived with only the most general assumptions about the nature of the isotropic distribution function,[7] and has also been extended to higher temperatures.[1] In this section we concentrate on ferromagnets, where the arguments are simplest and most rigorous.

Following Ref. 7, let ρ_m be the probability of a state in which an individual spin S has component m along the field axis. At sufficiently low temperatures only the states $m=S$ and $m=S-1$ are appreciably populated, so that

$$\langle Y_l^0\rangle_T = \langle S \mid Y_l^0 \mid S\rangle\rho_S/(\rho_S+\rho_{S-1}) + \langle S-1 \mid Y_l^0 \mid S-1\rangle\rho_{S-1}/(\rho_S+\rho_{S-1}).$$

The quantity $\langle S \mid Y_l^0 \mid S\rangle = \langle Y_l^0\rangle_0$, and both $\langle S \mid Y_l^0 \mid S\rangle$ and $\langle S-1 \mid Y_l^0 \mid S-1\rangle$ can be evaluated by the Wigner–Eckart theorem. The result is

$$\langle Y_l^0\rangle_T/\langle Y_l^0\rangle_0 = 1 - l(l+1)/2S[\rho_{S-1}/(\rho_S+\rho_{S-1})]. \tag{10}$$

Now the reduced magnetization is itself

$$m(T) = \langle Y_1^0\rangle_T/\langle Y_1^0\rangle_0 = 1 - S^{-1}\rho_{S-1}/(\rho_S+\rho_{S-1}). \tag{11}$$

Elimination of the probability factors in Eq. (11) in terms of the magnetization yields

$$\langle Y_l^0(\mathbf{S}_i)\rangle_T/\langle Y_l^0(\mathbf{S}_i)\rangle_0 \cong 1 - \tfrac{1}{2}[l(l+1)][1-m(T)] \cong m(T)^{l(l+1)/2}. \tag{12}$$

Extension to higher temperatures evolves from a recognition[8] that in many models (molecular field, spin wave, and at least two Green's function de-

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coupling schemes) the single-particle density matrix is approximated by

$$\rho_m = \exp[Xm]/\mathrm{tr}\exp[Xm]. \tag{13}$$

The symbol m designates the magnetic quantum number. The quantity X varies from model to model, being (magnetic moment)·(exchange field)/(kT) in the molecular field theory and more complicated things in other cases. The trick is to calculate both $\langle Y_l^0\rangle_T$ and $m(T)$, as in Eqs. (10) and (11), as functions of the unknown X, and eliminate X, thereby obtaining an almost model-independent relationship between statistical averages of different harmonics. Thus

$$\langle Y_l^0\rangle_T = \sum_{-S}^{S} \langle m \mid Y_l^0 \mid m\rangle \exp[Xm] / \sum_{-S}^{S} \exp[Xm] \tag{14}$$

and

$$Sm(T) = \sum_{-S}^{S} \langle m \mid Y_l^0 \mid m\rangle \exp[Xm] / \sum_{-S}^{S} \exp[Xm]. \tag{15}$$

Wolf[9] had already employed this technique within the molecular field model; the contribution of Ref. 8 was to recognize its greater validity.

For large spin values Eqs. (14) and (15) can be adequately replaced by classical continuous averages over angle. The result is

$$\langle Y_l^0\rangle_T/\langle Y_l^0\rangle_0 = I_{l+1/2}(X)/I_{1/2}(X)$$
$$= I_{l+1/2}[\mathfrak{L}^{-1}(m)]/I_{1/2}[\mathfrak{L}^{-1}(m)] \equiv \hat{I}_{l+1/2}\{\mathfrak{L}^{-1}[m(T,H)]\} \tag{16}$$

with

$$m(T,H) = \langle Y_1^0\rangle_T/\langle Y_1^0\rangle_0 = I_{3/2}(X)/I_{1/2}(X) = \mathfrak{L}(X). \tag{17}$$

$I_{l+1/2}$ is a reduced hyperbolic Bessel function. $\mathfrak{L}(X) \equiv I_{1/2}$ is the Langevin function. These functions are displayed graphically in Ref. 7.

Now let us look at the record. One of the most satisfying case histories is that of nickel. Although the magnetic anisotropy departs wildly from the above theory, varying something like the 50th power of the magnetization[10] (a power which is greatly reduced by the thermal expansion correction[10]), its strain derivative, the magnetostriction, can now be considered to have been tamed. Birss and Lee[11] measured the temperature dependence of λ_{100} and λ_{111} of nickel, the first of which actually displayed a broad maximum. Lee and Birss[12] fitted their data to a polynomial, $am^3+bm^{10}+cm^{21}$ $(l=2, 4, 6)$, which is not well motivated at higher temperatures. Callen and Callen[1] showed that the correction for the known temperature dependences of the elastic constants helped somewhat with λ_{100}, but that important disagreement remained. The paper further suggested that room-temperature measurements,[13] extrapolated by Eq. (16), showed that higher harmonics could not be ignored at lower temperatures. Tatsumoto, Okamoto, and Kadena[14] measured the five $l=2$ and $l=4$ λ coefficients of nickel, but did not compare with theory. Benninger and Pavlovic[15] finally went the full course. Although their data as taken look like anything but Eq. (16), they show that when transformations are made to the correct combinations of strains and the temperature dependences of the elastic constants are included, λ_{100} and λ_{111} do indeed follow the theory well. (That the $l=4$ terms do not is ascribed to the neglect of $l=6$ terms at low temperatures.) Lourens and Alberts[16] report on the forced component of λ_0, λ_{100}, and λ_{100} of Ni; their values are of the correct sign, and plausible, but there is no sufficiently accurate magnetization data by which their results can be precisely related to Eq. (16). Nakamichi and Yamamoto[17] have performed very thorough measurements of the saturation magnetostriction of nickel–manganese alloys. They have studied the effect of temperature, concentration, and atomic ordering near Ni_3Mn.

The behavior of Fe is more puzzling. Takaki,[18] Tatsumoto and Okamoto,[19] and Gersdorf, Stoelinga, and Rathenau[20] found that λ_{100} (but not λ_{111}) has a peak just below T_c, a peak which shifts to lower temperatures with silicon alloying. Callen and Callen[21] proposed two mechanisms, both of which depend upon an anisotropy in the magnon energy $\omega(\mathbf{q})$ as the direction of $\mathbf{q}$ varies with respect to the magnetization. In the presence of such an anisotropic surface of constant magnon energy, the Brillouin zone distorts to conform, the real lattice distorting contravariently, and, as this anisotropy depends upon $\mathbf{M}$, the distortion follows the magnetization, producing magnetostriction. One mechanism proposed is a dipolar interaction, of the opposite sign to the real dipolar energy and eight times

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as large. A more fashionable suggestion is anisotropic magnon–phonon coupling, with magnon renormalization accounting for the high-temperature peak. Both mechanisms yield the observed temperature dependences of λ_{100} and λ_{111}; the second also accounts for the shift with alloying of light ions. Neutron diffraction might be able to test these hypotheses. Belson,[22] probing the latter model, has measured λ_{100} on a nickel–cobalt crystal of the same Debye and Curie temperature as Fe, but observed no peak. He alloyed another light element, Al, in Fe, but observed no shift. On the other hand, cubic cobalt–iron displays a high-temperature peak. The present writer determinedly views the evidence as inconclusive. Lastly we mention Hasuo's report[23] of the room-temperature forced magnetostriction of the lowest six λ components of Fe. (Also see Ref. 3.)

There has been considerable interest in rare earths. Bozorth and Wakiyama[24] and Alstad and Legvold[25] measured the saturation and forced magnetostriction of Gd. Their data are in substantial agreement. Coleman and Pavlovic[5] report both saturation and forced terms, as do Belov, Yergin and Pedko.[26] In addition to the single-ion source which leads to Eq. (16), there can be two-ion terms in $\lambda^{\gamma,2}$. These are well approximated by $m^2(T, H)$. Reference 1 shows that $\lambda^{\gamma,2}$ of Gd is accurately represented by a reasonable linear combination of $\hat{I}_{5/2}$ and m^2.

The heavy rare earths display the largest known magnetostrictions, 1%, not only because degenerate ground states of the ions lead to large B coefficients (in fact, rare-earth ions show similarly large B coefficients when in garnets), but also because of the unusually small elastic constants (about five times softer than iron.) These large strains were first indicated by polycrystal measurements of Lee and Alberts[27] on Dy, and confirmed by x-ray measurements by Darnell and Moore,[28] and Darnell,[29] on the same element. Simultaneously, Legvold, Alstad, and Rhyne[30] and Clark, Bozorth and DeSavage[31] crashed Physical Review Letters with "giant magnetostriction". Finally Clark, DeSavage and Bozorth[32] presented conclusive data on Dy, in precise confirmation of Eq. (16). As the temperature was varied through the paramagnetic, antiferromagnetic spiral and ferromagnetic ranges, $\lambda^{\gamma}(T, H)$ changed by four decades, but followed the $\hat{I}_{5/2}$ law everywhere.

This statement requires qualification, because the theory of Eq. (16) applies only to ferromagnets. In the three rare earths Tb, Dy, and Ho the spins are constrained to lie near the basal plane. In one phase the spin structure is an antiferromagnetic spiral. This spiral can be suppressed by a magnetic field in the basal plane and the simple theory of ferromagnetic magnetostriction can then be applied. This is what was done on Dy, and on other rare earths discussed below. Some start has been made toward the analysis of the magnetostriction of more complex spin structures in the work of Lee[33] on basal plane spirals.

Although "giant magnetostriction" was reported on Dy, which was measured first, slightly larger strains are actually observed in Tb. This will be discussed in Sec. 4. Measurements on Tb have been reported by Darnell,[29] by Rhyne and Legvold,[34] who disagree with Darnell, and by DuPlessis and Albert,[35] who confirm the work of Rhyne and Legvold, which appears definitive. Rhyne and Legvold measure $l=2$ and $l=4$ coefficients and show that they agree remarkably with Eq. (16). In materials of such large anisotropy many coefficients cannot be measured in the magnetically ordered state. DeSavage and Clark[36,37] have measured coefficients of Eq. (2) in the paramagnetic region, shown that they follow the theoretical T and H dependence, and thereby extrapolated their values into the low-temperature region where they cannot be determined directly. In the paramagnetic region Eq. (16) becomes[1]

$$\hat{I}_{l+1/2} \rightarrow m^l(T, H) \sim \chi^l(T) H^l. \tag{18}$$

Rhyne, Legvold, and Rodine[30,38] report the change in length along principal crystallographic axes in holmium.

Erbium is an interesting case. Here the spins lie uniformly on a cone around the c axis. A large field in the basal plane collapses the cone to a fan, with the spins tipped at the same angle to the c axis. A larger field collapses the fan, so that all spins are parallel, but remaining on the original cone. Still larger fields rotate the spins into the basal plane. Rhyne and Legvold[39] have measured $\delta l/l$ along principal axes in Er. They relate the strains to ferromagnetic components of

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magnetization. In one phase, Er has an alternating sinusoidal magnetization component along the c axis. Unpublished calculations by Dimmock, Tsuya, and the author suggest that from strain modulation of both the indirect exchange interaction and one-ion anisotropy energy an oscillatory c axis strain should accompany the sinusoidal magnetization. This sinusoidal $\lambda^a \sim 5\times10^{-4}$ averages to zero under a strain gage, but may be marginally detectable as a pair of x-ray satellites. Much work remains to be done on the magnetostriction of complex spin structures.

Magnetostriction has not yet been measured on single-crystal Tm, because of difficulties in growing a single-crystal sample. This material should have the largest negative magnetostriction (Tb and Dy are the largest, but positive), according to calculations to be discussed in the next Section.

4. MAGNETOELASTIC CONSTANTS

Having discussed the simplest case, isotropic exchange and single-ion perturbations in ferromagnets, we now aim toward the more general Hamiltonian of Eq. (5), and multi-sublattice systems. To do so we must sketch the origins of the magnetoelastic coefficients themselves. Fortunately there are three excellent reviews. A detailed, insightful article by Kanamori[40] discussed the extensive original calculations of Tsuya[41] on $3d$ ions in spinels, applied by Slonczewski[42] to cobalt ferrite. Comstock[43] reviews, very readably, the origin, measurement methods and known magnetoelastic coefficients of spinels and garnets. Jones[44] amasses data on iron group and lanthanide ions up to 1966; later results will be referenced in due course.

A fundamental mechanism is a change in crystal field with strain. For $3d$ ions this can be combined either with $\lambda\mathbf{L}\cdot\mathbf{S}$ twice or with intra-atomic spin–spin coupling once. Generally the temperature dependence is complicated, depending upon the excitation of unequally spaced levels. For example λ_{100} and λ_{111} need not have the same variation with temperature. Perhaps the most comprehensive calculation of the magnetostriction of a $3d$ ion is that of Slonczewski,[42] but the observed temperature dependence does not agree with his predictions.[45]

Tsuya *et al.* have extended this approach to the rare-earth garnets[46] and also to metals.[47] In rare-earth metals conduction electrons play several important roles. The exchange interaction is isotropic, and comes from conduction-electron-spin polarization (RKKY). Conduction electrons screen the crystal field, so that $J^0 > D^0$ [Eq. (5)], the levels are equally spaced, the Callen–Shtrikman[8] theorem applies, and the $\hat{I}_{l+1/2}$ behavior follows, as we have already seen. Tsuya, Clark, and Bozorth[47] show that when the crystal field is changed by strain, conduction electrons redistribute, and this is the largest source of magnetostriction in heavy rare-earth metals.

Consider the anisotropic $4f$ charge cloud, with its symmetry axis along the magnetization. The change of crystal field accompanying distortion causes a change in electrostatic energy of the $4f$ electrons, which change depends upon magnetization direction. Suppose an oblate charge cloud in the yz plane has its orbital angular momentum along x in the basal plane. Neighboring + ions along y are attracted inward and + ions along x tend to move away (no volume strain) to minimize the electrostatic energy between $4f$ electrons and neighboring cores. The eccentricity of the charge density, reflected in the Stevens factor, changes from oblate to prolate between holmium and erbium, and accordingly the anisotropy and magnetostriction change sign. Conduction-electron redistribution accompanies the lattice distortion. The overriding contribution to the large magnetoelastic coefficients is from the change in electrostatic energy between conduction and $4f$ electrons on the same atom.[47]

DeSavage and Clark[36,37] measured the four $l=2$ anisotropic magnetostrictions of Tb in the paramagnetic phase, extrapolated by Eq. (16) and (18) to the 0°K values, and compared these to their calculations of λ. They show that the ion core contribution is greatly reduced by screening, and that some coefficients would have the wrong sign from crystal field alone. But the conduction electron redistribution effect is much larger, and produces λ coefficients in good agreement with the extrapolated measurements.

In an interesting recent paper on the origin of magnetostriction of $3d$ metals (see Secs. 2 and 3 for some others), Berger[48] relates a maximum in the magnetoresistance, a reversal in sign of the extraordinary Hall effect, and a change in sign of the magnetostriction of Fe–Ni and Co–Ni alloys, all at the same electron concentration, to an orbital degeneracy near the Fermi level. The degeneracy is lifted by spin orbit coupling, one resulting linear combination of Bloch states producing positive λ, the other negative.

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[47] N. Tsuya, A. E. Clark, and R. M. Bozorth, *Proceedings of the International Conference on Magnetism* (Institute of Physics and the Physical Society, London, 1965), p. 250.
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As the Fermi level shifts through the degeneracy upon alloying, the magnetostriction changes sign.

A phenomenological approach has been vigorously exploited in papers by Phillips and White on the magnetostriction of rare-earth iron garnets[49] and of iron-group monoxides,[50] on the basis of a quasi single-ion model. The distinction between "single-ion" and "two-ion" sources softens in multi-sublattice systems, and melts for rare-earth ions in garnets, in which the rare earth is only weakly exchange coupled to the strongly interlocked iron sublattices. Terms in the Hamiltonian like $\mathbf{S}_{\mathrm{Fe}}\cdot\mathbf{J}_{\mathrm{FeR}}\cdot\mathbf{S}_{\mathrm{R}}$ behave quite accurately like $\langle\mathbf{S}_{\mathrm{Fe}}\rangle\cdot\mathbf{J}_{\mathrm{FeR}}\cdot\mathbf{S}_{\mathrm{R}}$, a single-ion effective field energy, but of *odd* order in the rare earth spin operator. The anisotropic character of $\mathbf{J}$, or of $\partial\mathbf{J}/\partial\epsilon$, and the order ($l=1$) of the spin operator lead to new temperature and field dependence. Both the isotropic and longitudinal two-ion correlation functions of Eq. (6) reduce to $M_{\mathrm{Fe}}(T)M_{\mathrm{R}}(T)$. In the garnets there are six distinct, differently oriented rare-earth sites in the unit cell. This, with anisotropic exchange, produces a different set of splittings at many sites, which may each be unevenly spaced. But in some circumstances Eq. (16) can yet be generalized to apply. In any case, the magnetostriction is observed to be "single-ion" in some spinels[45] (but not all[51]) and garnets[52] in the sense that it is linear in concentration.

Phillips and White begin with Eq. (5). $\mathbf{J}^0$ and $\mathbf{D}^0$ are known. For Kramers doublet ($S_{\mathrm{eff}}=\frac{1}{2}$) ions such as Yb^{3+} (Er^{3+} is not a well separated Kramers doublet!) in RIG, $\mathbf{D}\equiv 0$, so that all effects must be related to variation of $\mathbf{J}$ with strain. Because $\partial\mathbf{J}/\partial\epsilon_\mu$ is presently unknown, but there is experimental data on the more-easily-measured paramagnetic g tensor, $\partial\mathbf{g}/\partial\epsilon$, the authors make an identification

$$\langle\mathbf{S}_{\mathrm{Fe}}\rangle\cdot\mathbf{J}_{\mathrm{FeR}}\cdot\mathbf{S}_{\mathrm{R}}\equiv\boldsymbol{\alpha}\cdot\mathbf{G}\cdot\mathbf{S}_{\mathrm{R}}=\mu_g\mathbf{H}_{\mathrm{eff}}\cdot\mathbf{g}\cdot\mathbf{S}_{\mathrm{R}}. \quad (19)$$

The required matrix $\partial\mathbf{J}/\partial\epsilon$ is then assumed proportional to $\partial\mathbf{g}/\partial\epsilon$, which misses the $(\partial H_{\mathrm{eff}}/\partial\epsilon)\mathbf{g}$ part of $\partial\mathbf{G}/\partial\epsilon$. $\partial\mathbf{g}/\partial\epsilon$ (and $\partial\mathbf{D}/\partial\epsilon$) is then taken from EPR measurements of the same rare-earth ion in a diamagnetic garnet host.

5. FERRIMAGNETS

The analysis of Sec. 3 presupposed a ferromagnetic array, and its experimental verification was demonstrated in nickel[15] and in rare earths in a ferromagnetically aligning field.[32,34] To apply the concepts of Sec. 4 we must now generalize to a multi-sublattice system, but we shall restrict our attention to Néel colinear arrays. Rigorously, the problem is not trivial because of the zero point sublattice reduction. Note that in Eq. (12) the transition from the middle to the last equality requires that

$$m(T)\underset{T\to 0}{\longrightarrow}1.$$

Nevertheless, for large spins and except at the lowest temperatures, a useful and accurate approximation is that

$$\lambda^{\mu,l}(T,H)=\sum_n\lambda^{\mu,l}(n)\hat{I}_{l+1/2}\{\mathfrak{L}^{-1}[m_n(T,H)]\}. \quad (20)$$

That is, the contribution to λ is additive over all sublattices n. Each sublattice has its own set of magnetoelastic constants $B_l^\mu(n)$, and all compete to pick that uniform strain which minimizes the total free energy. Of course Eq. (20) is subject to the same restrictions as Eq. (16); it applies only when the exchange is isotropic and the magnetostriction comes from $\mathbf{D}$, not $\mathbf{J}$ terms. Application of Eq. (20) requires a knowledge of all sublattice reduced magnetizations $m_n(T,H)$, and for each term in the magnetostriction, a set of magnetoelastic constants, one for each sublattice. Equation (20) was first proposed and applied[53] to YIG, which has two iron sublattices. Agreement with experiment is convincing, but then there are two adjustable B constants for each of λ_{100} and λ_{111}.

When the $B_l^\mu(n)$ on different crystallographic sites are of opposite signs, competition between sublattices can lead to complex thermal behavior of the net $\lambda^{\mu,l}(T,H)$. This occurs for Fe^{3+} ions on the octahedral and tetrahedral sites in both spinels and garnets. When the coefficient of larger magnitude is associated with the sublattice magnetization which drops off more rapidly with increasing temperature, a magnetostriction compensation temperature and reversal of sign of $\lambda(T)$ can ensue.[54] When the $\lambda^{\mu,l}(n)$ coefficient of smaller magnitude is associated with the more rapidly falling sublattice moment, $\lambda(T)$ can display a maximum at nonzero temperature.[53]

The magnetostriction compensation temperature is not simply related to the magnetization compensation temperature, the former coming from the competition of sublattice magnetostrictions, and depending upon magnetoelastic coefficients. Actually an anomaly is also observed at a magnetization compensation point,[46,52,55] more of observational than fundamental origin. Magnetostriction is measured by determining the change in length in some fixed direction $\boldsymbol{\beta}$ when the magnetization is rotated by an external field from one direction $\boldsymbol{\alpha}_i$ to another, $\boldsymbol{\alpha}_f$. But at the magnetization compensation point, where there is no net moment, the sublattice moments fail to follow the applied field,

[49] T. G. Phillips and R. L. White, Phys. Rev. Letters **16,** 650 (1966); J. Appl. Phys. **38,** 1222 (1967); Phys. Rev. **160,** 316 (1967).

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[51] N. Miyata and Z. Funatogawa, Proc. Intern. Conf. on Magnetism and Crystallography, Kyoto, J. Phys. Soc. Japan **17,** Suppl. B-1, 279 (1962).

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[54] A. E. Clark, B. F. DeSavage, and E. R. Callen, J. Appl. Phys. **35,** 1028 (1964).

[55] D. F. Bleil and A. R. Butz, Phys. Rev. **94,** 1440 (1954).

and for small fields, there is a dip in the *apparent* magnetostriction over a narrow temperature interval, beyond which it returns to its true value.

There is also a reversal in sign of the forced magnetostriction at the magnetization compensation temperature.[52] In traversing the compensation temperature all sublattice magnetizations flop over in the external field. If a sublattice magnetization is increased by the external field on one side of the compensation point, it is reduced by the field on the other; the forced moment is linear in H. Thus that part of the magnetostriction of ferrimagnets which is linear in H in the ordered phase, the forced magnetostriction, changes sign abruptly at the magnetization compensation temperature.

The previous discussion is valid for colinear arrays, with the sublattices aligned with the external field. This will not be the case unless the applied field is small compared to the inter-sublattice exchange field. The danger of nonalignment, and of angles between sublattice magnetizations is particularly acute near the compensation temperature, as we have seen. In TbIG complex $\lambda(R, H)$ have been reported,[56] which reflect the intricate orientations assumed by the several sublattices as the external field is rotated. Even for constant field, as the temperature is changed the net moment can rise, fall, remain constant or all three, because of sublattice rotation.[57] We now review the literature, first on spinels, then on garnets.

It was previously mentioned that the magnetostriction of $Co_xMn_{1-x}Fe_2O_4$, observed by Lee and Robey,[45] is linear in x. Miyata and Funatogawa[51] do not find such additivity in $Mn_xFe_{3-x}O_4$; on the contrary λ_{100} has a maximum at $x=0.6$, as does the magnetic anisotropy. The present author believes these maxima to be related to cation distributions.[58] Syono and Ishikawa[59] demonstrate the importance of Fe^{2+} on tetrahedral sites in $Ti_xFe_{3-x}O_4$. Smith and Jones[60] also emphasize the ferrous ion in their study of $NiFe_2O_4$ (which should not have any). They find λ_{100}, λ_{111}, g, K and linewidth all to be essentially temperature independent below room temperature. In $Co_{0.05}Ni_{0.95}Fe_2O_4$, λ_{100} remains fairly constant, but λ_{111} has a sharp temperature dependence. De Lacheisserie[61] examines the system $Zn{:}Ni_{1-x}Fe_2O_4$. He concludes that the polycrystal averages $\lambda_s(x, R)$ are consistent with additivity and Eq. (20). Ohta and Kobuyashi[62] have studied the temperature dependence of magnetic anisotropy and of λ_{100} and λ_{111} over the ternary field $(MnO)_x(ZnO)_y(Fe_2O_3)_z$, in order to find regions of $\lambda_s=K=0$, and a high-permeability ceramic ferrite.

This review is concerned with macroscopic strains, but we mention a unique study of internal magnetostriction. By means of x-ray diffraction Goldberg and McCann[63] observe a spatially uniform "breathing" of the oxygen u parameter as the magnetization is rotated.

The magnetostriction of rare-earth iron garnets is accurately linear in rare-earth concentration.[52] We can therefore subtract out the magnetostriction and the magnetization of the YIG background, and relate the rare-earth magnetostriction contribution to the rare-earth magnetization. More generally, when the exchange interaction is anisotropic, as is usually very much the case, we can consider the rare-earth magnetostriction as the sum of contributions from the six sublattices (for symmetry directions of the net magnetization, 2, 3, or 4 of the 6 will be equivalent), and relate each such sublattice magnetostriction to its own magnetization.

The simplest case to begin with is the S state ion, Gd^{3+}, of which the exchange coupling J^0 to the iron system is isotropic, so that we may consider all rare-earth sites simultaneously. Furthermore, for an S state ion we expect $\mathsf{J}^0 \gg \mathsf{D}^0$, so that the energy levels of the effective spin should be evenly spaced, and the Callen–Shtrikman approach should be valid. The question then is what is the perturbation: the exchange term $\partial\mathsf{J}/\partial\epsilon$, single-ion anisotropy $\partial\mathsf{D}/\partial\epsilon$, or both? Phillips and White,[49,64] who ascribe the single-ion source point out that exchange modulation is the more difficult to monitor by EPR under stress, not only because the ferromagnetic $\partial\mathsf{G}/\partial\epsilon$ may not be given by the paramagnetic $\partial\mathsf{g}/\partial\epsilon$, as discussed after Eq. (19), but because very small changes in the experimentally measured g are multiplied up by the large exchange field factor. The observed temperature dependence of λ_{100} of GdIG answers this question[52] in favor of the exchange modulation. The results are

$$\frac{\lambda_{100}^{\mathrm{GdIG}}(77°)-\lambda_{100}^{\mathrm{YIG}}(77°)}{\lambda_{100}^{\mathrm{GdIG}}(4.2°)-\lambda_{100}^{\mathrm{YIG}}(4.2°)}=0.63$$

$$\frac{M^{\mathrm{GdIG}}(77°)-M^{\mathrm{YIG}}(77°)}{M^{\mathrm{GdIG}}(4.2°)-M^{\mathrm{YIG}}(4.2°)}=0.60$$

$$\frac{\lambda_{100}^{\mathrm{GdIG}}(296°)-\lambda_{100}^{\mathrm{YIG}}(296°)}{\lambda_{100}^{\mathrm{GdIG}}(4.2°)-\lambda_{100}^{\mathrm{YIG}}(4.2°)}=0.19$$

$$\frac{M^{\mathrm{GdIG}}(296°)-M^{\mathrm{YIG}}(296°)}{M^{\mathrm{GdIG}}(4.2°)-M^{\mathrm{YIG}}(4.2°)}=0.18$$

Rare earth magnetostriction is proportional to rare

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[59] Y. Syono and Y. Ishikawa, J. Phys. Soc. Japan **18**, 1231 (1963); **19**, 1752 (1964); Y. Syono, Japanese J. Geophys. **4**, 71 (1965).
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[64] R. L. White and T. G. Phillips, J. Appl. Phys. 39, 579 (1968), this issue.

earth magnetization. (The variation of iron d sublattice magnetization is small in this temperature range.) In subtracting magnetizations one must be careful about signs. The compensation temperature of GdIG$\sim$285°K. The above number array is inconsistent with the single-ion mechanism, for which $\Delta\lambda$ would vary something like $(\Delta m)^3$ to $(\Delta m)^2$.

Another simple case, also discussed by Phillips and White[49,64] is Yb^{3+} in YIG. Here the ground state is a well-separated Kramers doublet (effective $S=\frac{1}{2}$), hence $\mathsf{D}=0$, and only $\partial\mathsf{J}/\partial\epsilon$ can cause magnetostriction. However J is very anisotropic, so that different sites see different exchange splittings. For each site, $\lambda\sim M\sim\tanh(\Delta E/kT)$. Because the contributions of the six sites are weighted differently[65] in summing to find the total rare-earth λ and M (the $\mathbf{M}$ of the sites are not parallel) these totals need *not* be related by a proportionality constant independent of temperature, although this at times turns out to be pretty much so, because of averaging in summing over the six sites. Calculations by Alben[65] on YbIG show that $\sum\lambda/\sum M$ is approximately a constant, independent of temperature to within 5% above 40°K. Comstock and Raymond[66] have measured the magnetostriction of Yb^{3+} and of Ce^{3+} in YIG. For the former ion they report the behavior described above, and interpret it in the same way.

There have also been many other measurements of the magnetostriction of RIG. Jones[44] lists many macroscopic magnetoelastic coefficients, which are defined by $\lambda_{\text{total}}\sim B/c$ as in Eq. (6). Such macroscopic constants, related only in a highly derived fashion to the intrinsic site coefficients, are temperature-dependent quantities. Iida[67] measured $\lambda_{100}(T)$ and $\lambda_{111}(T)$ of fully substituted EuIG and TbIG above 77°K, where the rare earth is largely disordered. He reports that the second-order ΔK due to magnetostriction ($\Delta K\sim B^2/c$) is larger than the intrinsic K of the clamped crystal. Smith and Jones[68] introduced a technique similar to that of White, but where White performs EPR under pressure, in diamagnetic hosts, Jones measures ferromagnetic resonance shifts with pressure directly in the magnetic host. Jones measures $\partial\mathsf{J}/\partial\epsilon$ and $\partial\mathsf{D}/\partial\epsilon$ directly, whereas EPR misses the strain dependence of the exchange field. Smith and Jones studied YIG, and YIG doped with Er^{3+} and Yb^{3+}. The NOL group have reported on YIG,[53] GdIG,[52] DyIG[46,52] HoIG,[46] and ErIG.[46] Some recent results are tantalizing but not yet understood: for DyIG, $\lambda_{111}(T)$ follows the $\hat{I}_{5/2}$ law very well but $\lambda_{100}(T)$ does not. Flanders, Pearson and Page[69] measure λ_{100} and λ_{111} at helium, nitrogen, and room temperature, on YIG, 10% Yb^{3+} and Tb^{3+} doped YIG, and on TbIG. Iida[67] reports λ_{100} and λ_{111} at 78°K, 196°K and room temperature on fully substituted garnets of all rare earths from Sm to Yb. Bertram and Jones,[71] by pressure-induced shifts in ferromagnetic resonance frequency, relate magnetoelastic coupling to $\partial\mathsf{J}/\partial\epsilon$, in $Eu_{3-x}Y_xFe_{5-y}Ga_yO_{12}$ (mixed iron–gallium garnets). The Eu^{3+} ion has a $J=0$ ground state, but $J=1$ states are admixed by the iron exchange field.

The last ferrimagnetic system to be mentioned, not a garnet, is Fe_5Ge_3. Tawara[72] constructs a two-sublattice molecular field model which reproduces his observed net $M(T)$ and $K(T)$, and he reports room-temperature magnetostrictions.

6. ANTIFERROMAGNETS

Magnetostriction of antiferromagnets faces difficulties which parallel but transcend the problem of magnetic anisotropy of such materials. Some workers have carried over Eq. (1) by interpreting $\boldsymbol{\alpha}$ as the direction of the external field, but the λ coefficients are then very much functions of external field strength but only dim images of microscopic patterns. A more revealing approach, which has apparently not been pursued, is to recognize additivity of sublattice contributions and write, for example,

$$\delta l/l=\lambda_{100}(T,H)\{[\alpha_1^2(1)+\alpha_1^2(2)]\beta_1^2+[\alpha_2^2(1)+\alpha_2^2(2)]\beta_2^2+[\alpha_3^2(1)+\alpha_3^2(2)]\beta_3^2\}+\text{other terms.}\quad(21)$$

Sublattice $\boldsymbol{\alpha}(i)$ are to be related to the external field by minimization of the free energy, including Zeeman, anisotropy and exchange. The field dependence of $\lambda_{100}(T,H)$ so defined is now only the usual forced magnetostriction from a sublattice paraprocess, rather than from rotation of sublattices.

Little experimental work has been done on the magnetostriction of antiferromagnets, primarily because of their irresponsiveness to external fields and difficulties in eliminating domain effects. Several papers,[73–75] notably Alberts and Lee,[73] served to establish that below saturation the observed strain varies as H^2 because of displacement of domain walls between spins in $\langle 111\rangle$ planes in iron-group monoxides. By a group-theoretical argument Birss and Anderson[76] established that in thirty-five magnetic crystal classes the forced

[65] Richard Alben and Earl Callen (to be published).
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magnetostriction beyond saturation should vary as H (as in ferromagnets) rather than the H^2 dependence of the more common antiferromagnets of higher symmetry. Suppose the anisotropy energy to be sufficiently large that the field can lie along the common axis $\alpha_{(1)}-\alpha_{(2)}$. Increasing H decreases the magnetization of one sublattice to the same extent that it increases that of the other, cancelling the linear term in total λ', in Eq. (8); the first net forced term is then $\lambda''H^2$. Only when the symmetry is so low that the field affects the two sublattices differently is $\lambda'H$ present. Anderson, Birss and Scott[77] took advantage of the small parasitic moment of α-Fe_2O_3 to confirm their earlier prediction. Iida and Tasaki[78] also employed α-Fe_2O_3 to investigate magnetoelastic properties of an antiferromagnet. A novel effect they demonstrate involves the rotation of the spin axis vector in the basal plane, accompanied by magnetostrictive tipping of the c axis toward the plane. As the spin vector rotates in one sense, the tipped c axis precesses in the opposite sense at twice the frequency. Haefner, Stout and Barrett[79] have examined the lattice parameters of another weak ferromagnet, NiF_2, by x-ray diffraction. Magnetic anisotropy of antiferromagnets is measured not by torque methods but by susceptibility, spin flop field, and resonance. One wonders whether antiferromagnetic resonance under uniaxial stress, *a la* Jones,[60] can be employed to advantage to determine magnetoelastic constants.

The theory has suffered from a paucity of experiments. Kanamori[40] reviews calculations of exchange-motivated lattice deformations of iron-group monoxides, calculations of which Kanamori and Nagamiya are principal authors. Phillips and White[50] employ their EPR technique on the same materials, but conclude that the source of magnetostriction is from single-ion anisotropy, $\partial \mathbf{D}/\partial\epsilon$, in NiO and MnO, rather than the $\partial \mathbf{J}/\partial\epsilon$ source calculated by Kanamori. This may perhaps be due to the same difficulty that arose in connection[49] with GdIG, namely inferring $\partial \mathbf{J}/\partial\epsilon$ from the very small paramagnetic $\partial \mathbf{g}/\partial\epsilon$ (see Sec. 5).

7. OSCILLATORY MAGNETOSTRICTION OF DIAMAGNETS

Although it does not properly belong in this review, the author cannot refrain from a brief discussion of oscillations in length ($\delta l/l \sim 10^{-8}$) of diamagnets, of de Haas–van Alphen origin, passage of Landau levels through the Fermi surface.[80] The free energy is [81]

$$F = 2kT \sum_{s=1}^{\infty} (-1)^s \left(\frac{eB}{2\pi sc\hbar}\right)^{3/2} \frac{[S'']^{-1/2}}{\sinh[2\pi^2 skT/\hbar\omega_c]} \cdot \cos[(sc\hbar/eB)\,S \pm \tfrac{1}{4}\pi] + \tfrac{1}{2}c_\mu \epsilon_\mu^2. \quad (22)$$

S is an extremal cross section of the Fermi surface normal to $\mathbf{B}$. It can vary with strain:

$$S = S_0 + \epsilon_\mu (\partial S/\partial \epsilon_\mu) + \cdots. \quad (23)$$

Often the harmonics s are experimentally well separated, or can be Fourier resolved. Keeping only the lowest harmonic for simplicity, and the most strain-sensitive factor and expanding for small strains,

$$F = -A\epsilon_\mu \sin(c\hbar S_0/eB \pm \tfrac{1}{4}\pi) + \tfrac{1}{2}c_\mu \epsilon_\mu^2, \quad (24)$$

and the equilibrium strain is

$$\epsilon_\mu = (A/c_\mu)\, \sin(c\hbar S_0/eB \pm \pi/4). \quad (25)$$

Hence ϵ_μ oscillates in $1/B$ with a period which measures the cross sectional area of the Fermi surface.[82–84] The amplitudes of λ_0, λ_{100} and λ_{111} oscillations can be related to deformation potentials and distortions of the Fermi surface. Interesting complications arise when $B(\epsilon) = H + 4\pi M(\epsilon)$ oscillates significantly (dHvA).

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"I played there were no deeper seas,
Nor any wider plains than these, ···.

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[78] S. Iida and A. Tasaki, *Proceedings of the International Conference on Magnetism* (Institute of Physics and the Physical Society, London, 1965), p. 583.
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