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Static Magnetoelastic Coupling in Cubic Crystals*

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The static magnetoelastic coupling in ferromagnetic or antiferromagnetic cubic crystals is analyzed in terms of a general formalism dictated by symmetry considerations. Besides the coupling of the spins to the external strains, resulting in external magnetostriction, the spins can also couple to internal strain modes. Only particular types of ionic displacements can couple to the spins, and these are classified. The spin operators which enter the theory are analyzed in terms of Tensor Kubic Operators, which are operator analogs of the Kubic harmonics, and which generate the irreducible representations of the cubic group. All equilibrium ionic displacements are found explicitly, and their temperature dependence is obtained. These equilibrium strains then lead to a general expression for the magnetoelastic contribution to the anisotropy energy and to the specific heat. On the basis of the usual $l(l+1)/2$ power law we derive the temperature dependence of the magnetoelastic coupling coefficients and of their contributions to the anisotropy energy and specific heat. The available experimental data on magnetostriction, magnetization, and elastic constants for nickel are specifically analyzed. In general, magnetically induced strains lower the symmetry from cubic, depending on the direction of the magnetization and on the particular strain modes supported by the crystal. We analyze these deviations from cubic symmetry and show which symmetry groups remain below the magnetic transition.

1. INTRODUCTION

By virtue of the dependence on distance of the exchange integral, of the spin-orbit interaction, or of the dipole-dipole interaction, the spin system in a ferromagnetic or antiferromagnetic crystal is coupled to the ionic displacements. The static portion of this interaction results in a shift in the equilibrium ionic positions (relative to the case with no magnetoelastic coupling), with resultant shifts of both the phonon and magnon spectra. The dynamic portion of the interaction produces magnon-phonon scattering.

The simplest aspect of the static interaction, and the aspect which has been considered previously, is the external magnetostriction, or the change in the macroscopic crystal dimensions. In addition there are shifts in the ionic coordinates within each unit cell, and, in some circumstances, this "internal" magnetostrictive coupling may be considerably larger than the external magnetostriction. Furthermore, the induced ionic displacements modify the symmetry of the crystal and reflect back to alter the magnetic properties, possibly changing the nature of the Curie transition (from second order to first order), and changing the temperature dependence of the anisotropy energy. In most common materials this alteration is small, but, again, there are circumstances in which it can be relatively large and significant. We shall, here, develop a general theory of the magnetoelastic coupling, including all types of elastic modes (which we classify according to their group theoretical properties), and considering explicitly the influence of this coupling on the magnetic properties. In addition, in Sec. 8, we will classify the possible crystal symmetries which, by virtue of the magnetoelastic coupling, can appear below the Curie temperature in a crystal which is cubic above the Curie temperature.

Direct observation of the internal magnetostriction is, unfortunately, more difficult than observation of the external magnetostriction. However, x-ray observations may detect some shifts, such as that of the oxygen "*u* parameter" of ferrospinels, which are elastically "soft." The longitudinal standing spin waves and canted spin arrangements of the rare earths, of hausmannite, and of various other ferrimagnets should also produce characteristic internal strains or, in certain cases, superlattice lines which may be observable by x-ray means. But perhaps the most sensitive way to observe the magnetostrictive coupling to internal modes is by resonance. In particular, rotation of the magnetization alters the ionic positions within the unit cell and changes the crystalline fields and orbital overlaps. These alterations should be detectable as shifts in nuclear resonance frequencies. Another possibility is that the spin-lattice interaction (and, hence, the ferrimagnetic resonance linewidth) of the rare-earth ions in doped garnets may reflect the shift of internal ions with rotation of the magnetization. Furthermore, the destruction of the tenth-power law for the magnetocrystalline anisotropy, alteration of the temperature dependence of the external magnetostriction, a change in the type of magnetic phase transition from second order to first order, and an anisotropic contribution to the specific heat, can all provide observational evidence for internal magnetostrictive coupling.

The classical static theory of magnetostriction in cubic crystals was originally given by Becker and Döring.[1] In that theory the magnetization is coupled

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[1] R. Becker and W. Döring, *Ferromagnetismus* (Verlag Julius Springer, Berlin, 1939), p. 132, 145.

to the uniform macroscopic strain by terms in the free energy, involving various polynomials in the strains multiplied by polynomials in the direction cosines of the magnetization. The form of these polynomials is dictated entirely by symmetry considerations, and the magnitude of these coupling terms is represented by phenomenological magnetoelastic coupling coefficients, of unknown temperature dependence.

We follow a similar, but quantum-mechanical procedure, coupling the spin and elastic modes in the Hamiltonian rather than in the free energy with temperature-independent coupling coefficients. This requires consideration of the symmetry of spin-operator functions, and the introduction of those combinations of spherical tensor operators which generate the irreducible representations of the cubic group. These operators are the quantum mechanical analogs of the Kubic harmonics of Von der Lage and Bethe,[2] and we shall refer to them as Tensor Kubic Operators (TKO). The requirement that the Hamiltonian be fully symmetric under all the operations of the cubic group then dictates the form of the magnetoelastic interaction, as well as of the elastic energy. We shall find that in cubic crystals there are only five characteristic types of magnetoelastic coupling terms, three of which appear in the external dilatations and shears as well as in the internal modes. Only certain symmetry classes of modes can couple to the spins. For each of the five types of coupling terms, we calculate the equilibrium strains as a function of temperature and of magnetization direction, the resultant crystal symmetry, and the contribution of these terms to the magnetocrystalline anisotropy energy and to the specific heat.

For the sake of analytic simplicity we restrict our treatment to those cases in which the magnetoelastic coupling is the sum of interactions of single spins with the strain field (excluding, for instance, magnetoelastic coupling arising from a strain dependence of the exchange integral). Particularly in the antiferromagnetic oxides the dominance of the one-ion source of the coupling is strongly suggested by the one-ion character of the magnetocrystalline anisotropy, as suggested in ferrites and garnets by Yosida and Tachiki,[3] and by Wolf,[4] and demonstrated by Folen and Rado[5] and by Geschwind.[6]

We further restrict our treatment to those structures in which all magnetic ions are crystallographically equivalent and in which their average spin directions are all coaxial; again, this applies to most simple ferromagnetics and antiferromagnets, although canted and spiral spin structures are excluded. Generalization to these more complex structures will be given elsewhere.

Although the internal magnetostriction can destroy the tenth-power law for the temperature dependence of the anisotropy in a ferromagnet, introducing both low-power terms and other terms varying as very high powers, it should be noted that these terms cannot account for the puzzling behavior of iron[7] and nickel[8] at very low temperatures. (It should be recognized though that over most of the range of magnetization, the classical theory describes the temperature dependence of the anisotropy rather well.) These metals have only a single ion per unit cell, and consequently possess no internal modes of homogeneous ionic displacement. Furthermore, the coefficients of the magnetoelastic coupling to the external strain modes are known from magnetostriction measurements and are too small to produce the observed deviations of the temperature dependence of the anisotropy. However, the temperature dependence of the magnetostriction is fairly well accounted for by the theory, as we demonstrate in Sec. 7 by examination of the available data.

2. THE GENERAL HAMILTONIAN

The Hamiltonian is

$$H=H_m+H_e+H_{me}+H_a, \tag{1}$$

these terms being the intrinsic magnetic energy, the elastic energy, the magnetoelastic coupling, and the intrinsic anisotropy energy, respectively. The interaction with an external field, if present, is included in H_m; it will be reflected in the analysis by the specification of a direction of the average magnetization, $\boldsymbol{\zeta}$.

The magnetoelastic energy, which must transform according to the fully symmetric irreducible representation Γ_α, is to be formed from the direct product of the spin and elastic basis functions. As the direct product of two representations contains Γ_α only if the representations are equivalent, we can immediately limit the representations which are permissible.

Consider first the effect of time reversal, which is an element of the cubic group (although not of the full Shubnikov group of the magnetic crystal). Under time reversal, all strain components are invariant, whereas spin components are reversed. Hence, real TKO's involving odd powers of the spin operators transform under irreducible representations which are antisymmetric in the time reversal, and such TKO's cannot couple with the elastic components. We therefore restrict ourselves to real TKO's of even degree.

Conversely, we limit the permissible strain modes by considering the operation of spatial inversion. Every spin component, and, hence, every TKO, is invariant under this operation. Consequently the spins can couple

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[5] V. J. Folen and G. T. Rado, J. Appl. Phys. **29**, 438 (1958).

[6] S. Geschwind, Phys. Rev. **121**, 363 (1961).

[7] C. D. Graham, Jr., Fifth Conference on Magnetism and Magnetic Materials, November 16–19, 1959, Detroit, Michigan [J. Appl. Phys. (to be published)]. At low temperatures K_1, for iron, seems to vary as less than the fifth power of the magnetization.

[8] E. W. Pugh and B. E. Argyle, IBM Research Note NC-32 (unpublished). These authors report that the first anisotropy constant varies as rapidly as the 100th power of the magnetization.

TABLE I. The classical Kubic harmonics which are even under inversion. All functions normalized to unity. Functions in square brackets indicate functions with normalization factors omitted. Factors of ρ^{-l} are omitted throughout. From Von der Lage and Bethe (reference 2).

Γ_α	$K^{\alpha,0}(\zeta)=(4\pi)^{-1/2}$
	$K^{\alpha,4}(\zeta)=(4\pi)^{-1/2}[5(3\times7)^{1/2}/4]\{x^4+y^4+z^4-\frac{3}{5}\rho^4\}$
	$K^{\alpha,6}(\zeta)=(4\pi)^{-1/2}[3\times7\times11(2\times13)^{1/2}/8]\{x^2y^2z^2+(1/22)[K^{\alpha,4}]\rho^2-(1/105)\rho^6\}$
$\Gamma_{\beta'}$	$K^{\beta',6}(\zeta)=(4\pi)^{-1/2}[(2\times3\times5\times7\times11\times13)^{1,2}/8]\{x^4(y^2-z^2)+y^4(z^2-x^2)+z^4(x^2-y^2)\}$
Γ_γ	$K_1{}^{\gamma,2}(\zeta)=(4\pi)^{-1/2}(5)^{1/2}\{z^2-\frac{1}{2}(x^2+y^2)\}$
	$K_2{}^{\gamma,2}(\zeta)=(4\pi)^{-1/2}[(3\times5)^{1/2}/2]\{x^2-y^2\}$
	$K_1{}^{\gamma,4}(\zeta)=(4\pi)^{-1/2}[7(3\times5)^{1/2}/2]\{z^4-\frac{1}{2}(x^4+y^4)-(6/7)[K_1{}^{\gamma,2}]\rho^2\}$
	$K_2{}^{\gamma,4}(\zeta)=(4\pi)^{-1/2}[3\times7(5)^{1/2}/4]\{x^4-y^4-(6/7)[K_2{}^{\gamma,2}]\rho^2\}$
	$K_1{}^{\gamma,6}(\zeta)=(4\pi)^{-1/2}[11(2\times7\times13)^{1,2}/4]\{z^6-\frac{1}{2}(x^6+y^6)-(15/11)[K_1{}^{\gamma,4}]\rho^2-(5/7)[K_1{}^{\gamma,2}]\rho^4\}$
	$K_2{}^{\gamma,6}(\zeta)=(4\pi)^{-1/2}[11(2\times3\times7\times13)^{1/2}/8]\{x^6-y^6-(15/11)[K_2{}^{\gamma,4}]\rho^2-(5/7)[K_2{}^{\gamma,2}]\rho^4\}$
Γ_ϵ	$K_z{}^{\epsilon,2}(\zeta)=(4\pi)^{-1/2}(3\times5)^{1/2}xy$
	$K_z{}^{\epsilon,4}(\zeta)=(4\pi)^{-1/2}[3\times7(5)^{1/2}/2]xy\{z^2-(1/7)\rho^2\}$
	$K_z{}^{\epsilon,6}(\zeta)=(4\pi)^{-1/2}[3\times11(2\times3\times5\times7\times13)^{1/2}/16]xy\{z^4-(6/11)z^2\rho^2+(1/33)\rho^4\}$
	$K_z{}^{\epsilon,6'}(\zeta)=(4\pi)^{-1/2}[(2\times3\times7\times11\times13)^{1/2}/2]xy\{x^4+y^4-(5/8)(x^2+y^2)^2\}$
$\Gamma_{\delta'}$	$K_z{}^{\delta',4}(\zeta)=(4\pi)^{-1/2}[3(5\times7)^{1/2}/2]xy\{x^2-y^2\}$
	$K_z{}^{\delta',6}(\zeta)=(4\pi)^{-1/2}[3\times11(7\times13)^{1/2}/4]xy\{x^2-y^2\}\{z^2-(1/11)\rho^2\}$

only to strain modes which are symmetric under spatial inversion.

The irreducible representations of the cubic group are ten in number, of which only five are even under inversion. Following Von der Lage and Bethe, these five representations are denoted by Γ_α (the fully symmetric representation); $\Gamma_{\beta'}$ (one-dimensional); Γ_γ (two-dimensional); Γ_ϵ and $\Gamma_{\delta'}$ (both three-dimensional). The Kubic harmonics, which are classical basis functions for these representations, are given in Table I, which is taken from Von der Lage and Bethe with a modification in normalization; we prefer to normalize all functions to unity.

We now consider each of the separate terms in the Hamiltonian.

First, we construct the elastic Hamiltonian. The complete specification of the ionic configuration of the crystal is given by the standard strain components ϵ_{xx}, ϵ_{yy}, ϵ_{zz}, ϵ_{xy}, ϵ_{yz}, ϵ_{zx}, plus a number of additional coordinates specifying the displacements of the ions relative to the center of the unit cell. Linear combinations of these coordinates form bases for the irreducible representations. Thus, the six external strain components are replaced by the following six quantities:

$$\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz},\quad \text{belonging to } \Gamma_\alpha; \tag{2a}$$

$$5^{1/2}[\epsilon_{zz}-\tfrac{1}{2}(\epsilon_{xx}+\epsilon_{yy})],\quad \tfrac{1}{2}(15)^{1/2}[\epsilon_{xx}-\epsilon_{yy}],\quad \text{belonging to } \Gamma_\gamma; \tag{2b}$$

$$\epsilon_{yz},\ \epsilon_{zx},\ \epsilon_{xy},\quad \text{belonging to } \Gamma_\epsilon. \tag{2c}$$

[It will be noted that the symbol ϵ represents both the strains, and one of the irreducible representations. Both uses are conventional, and the ambiguity is resolved by the context.] Similarly, the internal coordinates are to be replaced by linear combinations of the proper symmetry.

Let the strain coordinates which belong to Γ_α be denoted by $\epsilon^{\alpha,j}$, with the value $j=0$ reserved for the volume dilatation

$$\epsilon^{\alpha,0}\equiv(\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz}). \tag{3a}$$

Similarly strain coordinates belonging to $\Gamma_{\beta'}$ are denoted by $\epsilon^{\beta',j}$, with j numbering the various coordinates of this type. There is no coordinate of this type with $j=0$ (i.e., no external strain).

Pairs of strain coordinates which generate the two-dimensional representation Γ_γ are denoted by $\epsilon_1{}^{\gamma,j}$ and $\epsilon_2{}^{\gamma,j}$, these components transforming like the Kubic harmonics $K_1{}^{\gamma,2}(\zeta)$, and $K_2{}^{\gamma,2}(\zeta)$, respectively. The external strains are again characterized by $j=0$;

$$\epsilon_1{}^{\gamma,0}\equiv5^{1/2}[\epsilon_{zz}-\tfrac{1}{2}(\epsilon_{xx}+\epsilon_{yy})], \tag{3b}$$

and

$$\epsilon_2{}^{\gamma,0}\equiv\tfrac{1}{2}(15)^{1/2}[\epsilon_{xx}-\epsilon_{yy}]. \tag{3c}$$

Finally, triplets of coordinates belong to Γ_ϵ are denoted by $\epsilon_i{}^{\epsilon,j}$ $(i=1,2,3)$ and those belonging to $\Gamma_{\delta'}$ are denoted by $\epsilon_i{}^{\delta',j}$ $(i=1,2,3)$. Only in the former case is the value $j=0$ present, with

$$\epsilon_1{}^{\epsilon,0}\equiv\epsilon_{yz},\quad \epsilon_2{}^{\epsilon,0}\equiv\epsilon_{zx},\quad \text{and}\quad \epsilon_3{}^{\epsilon,0}\equiv\epsilon_{xy}. \tag{3d}$$

The elastic energy, in the harmonic approximation, arises from the direct product of first-order strain representations. For the two-dimensional representations the fully symmetric quantity extracted from the direct product of the pair $\epsilon_1{}^{\gamma,j}\,\epsilon_2{}^{\gamma,j}$, and the pair $\epsilon_1{}^{\gamma,k}$, $\epsilon_2{}^{\gamma,k}$ is simply $\epsilon_1{}^{\gamma,j}\epsilon_1{}^{\gamma,k}+\epsilon_2{}^{\gamma,j}\epsilon_2{}^{\gamma,k}$. Similarly for the three-dimensional representations, the fully symmetric combination is $\sum_i\epsilon_i{}^{\epsilon,j}\epsilon_i{}^{\epsilon,k}$ (and similarly for $\Gamma_{\delta'}$). Therefore, the most general fully symmetric harmonic strain energy is

$$H_e=\tfrac{1}{2}\sum_{\mu}\sum_{j,k}c_{j,k}{}^{\mu}\sum_{i}\epsilon_i{}^{\mu,j}\epsilon_i{}^{\mu,k}, \tag{4}$$

or, if we assume the strain components to be chosen so as to diagonalize this quadratic form, the expression for the elastic energy becomes

$$H_e=\tfrac{1}{2}\textstyle\sum_{\mu}\sum_{j}c_j{}^{\mu}\sum_{i}(\epsilon_i{}^{\mu,j})^2. \tag{5}$$

We recall that j and k number the various modes of a given type of representation; μ takes the five values $\alpha, \beta', \gamma, \epsilon, \delta'$; and i takes only the single value unity for $\mu=\alpha$; and β' takes the values 1 and 2 for $\mu=\gamma$, and takes 1, 2, 3 for $\mu=\epsilon$ and δ'. The quantities $c_{j,k}{}^{\mu}$ (or $c_j{}^{\mu}$) are elastic constants.

For $j=0$ the elastic constants $c_0{}^{\mu}$ are related to the conventional elastic constants c_{11}, c_{12}, and c_{44} as follows:

$$c_0{}^{\alpha}=\tfrac{1}{3}(c_{11}+2c_{12}), \tag{6a}$$

$$c_0{}^{\gamma}=(2/15)(c_{11}-c_{12}), \tag{6b}$$

$$c_0{}^{\epsilon}=c_{44}. \tag{6c}$$

It will be noted that we have appended only a single subscript to the $c_0{}^{\mu}$, implying that the external strain modes $\epsilon_i{}^{\mu,0}$ are normal modes [compare Eqs. (4) and (5)]. In this matter we have two possibilities. If the $\epsilon_i{}^{\mu,0}$ are interpreted as the external strains as commonly measured (by a strain gauge for instance), they contain internal contributions which automatically admix so as to form a normal mode. The associated elastic constants are then the empirical elastic constants, as defined by Eq. (6). Direct empirical evidence for this admixture of internal displacements to the external strains has been given by Walsh[9] and by Kaminow and Jones,[10] who studied paramagnetic and ferrimagnetic resonance as a function of pressure. An alternative approach would be to define the external strain components, in accordance with elementary elasticity theory, as the coefficient of a homogeneous distortion; that is, all interatomic distances are proportionally increased, and the resulting strain is a linear combination of the true normal modes.

The magnetoelastic energy is obviously given in a completely analogous fashion. Thus,

$$H_{me}=-\sum_{\mu}\sum_{j,l}B_{j,l}{}^{\mu}\sum_{i}\epsilon_i{}^{\mu,j}\mathcal{K}_i{}^{\mu,l}. \tag{7}$$

The quantity $\mathcal{K}_i{}^{\mu,l}$ is a TKO belonging to the irreducible representation Γ_{μ}. If Γ_{μ} is three dimensional, $i=1, 2, 3$ and similarly for other dimensionalities. Again, l numbers the different possible TKO's which belong to the given representation. The constants $B_{j,l}{}^{\mu}$ are phenomenological magnetoelastic coupling coefficients.

As we have mentioned before, the TKO's are linear combinations of spherical tensor operators. As we shall see in the following section, TKO's of Γ_{α} cannot be formed from spherical tensors of degree 2, but can be formed from spherical tensors of degree 4 and 6. Similarly, there are TKO's of $\Gamma_{\beta'}$ of degree 6; of Γ_{γ} of degree 2, 4, 6; of $\Gamma_{\delta'}$ of degree 4 and 6; and of Γ_{ϵ} of degree 2, 4 and *two* different forms each of degree 6. We adopt the degree of the TKO as the labeling index l. Thus, l takes the following values:

$$\begin{aligned}
\mu&=\alpha, & l&=4, 6, \cdots;\\
\mu&=\beta', & l&=6, \cdots;\\
\mu&=\gamma, & l&=2, 4, 6, \cdots;\\
\mu&=\delta', & l&=4, 6, \cdots;\\
\mu&=\epsilon, & l&=2, 4, 6, 6', \cdots.
\end{aligned} \tag{8}$$

The last term in the Hamiltonian represents the intrinsic magnetic anisotropy of the unstrained crystal. It may be pseudodipolar or pseudoquadrupolar, or it may be a single-ion anisotropy arising from spin-orbit coupling and crystalline field splitting. In the latter case the anisotropy term in the Hamiltonian would have the particular form

$$H_a=\textstyle\sum_{l}\kappa_l\mathcal{K}^{\alpha,l}, \tag{9}$$

where κ_l is the "intrinsic anisotropy coefficient." For definiteness, we shall assume this form of the anisotropy, although this is in no way necessary. In fact, the Hamiltonian should contain additional terms involving the amplitudes of the phonons, or of the ionic oscillations relative to the average positions as described by the ϵ's. These include terms coupling spins to phonons, giving rise to dynamic aspects, or phonon-magnon scattering. The terms quadratic in the phonon amplitudes give a temperature-dependent elastic energy; if the phonon spectrum is dependent on the ϵ_i's, this effectively introduces a temperature dependence in the elastic constants. However, this temperature dependence is known to be small empirically.

Recapitulating, the total Hamiltonian is

$$\begin{aligned}H=H_m&+\tfrac{1}{2}\sum_{\mu}\sum_{j}c_j{}^{\mu}\sum_{i}(\epsilon_i{}^{\mu,j})^2\\
&-\sum_{\mu}\sum_{j,l}B_{j,l}{}^{\mu}\sum_{i}\epsilon_i{}^{\mu,j}\mathcal{K}_i{}^{\mu,l}+\sum_{l}\kappa_l\mathcal{K}^{\alpha,l}.\end{aligned} \tag{10}$$

3. THE FORMAL SOLUTION

Expanding to first order in the magnetoelastic coupling coefficients, the free energy is

$$\begin{aligned}F=F_m&+\tfrac{1}{2}\sum_{\mu}\sum_{j}c_j{}^{\mu}\sum_{i}(\epsilon_i{}^{\mu,j})^2\\
&-\sum_{\mu}\sum_{j,l}B_{j,l}{}^{\mu}\sum_{i}\epsilon_i{}^{\mu,j}\langle\mathcal{K}_i{}^{\mu,l}\rangle+\sum_{l}\kappa_l\langle\mathcal{K}^{\alpha,l}\rangle,\end{aligned} \tag{11}$$

where $\langle\ \rangle$ denotes a dynamical average with respect to the unperturbed density matrix.

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[9] W. M. Walsh, Jr, Phys. Rev. **114**, 1473, 1485 (1959).

[10] I. P. Kaminow and R. V. Jones, Phys. Rev. **123**, 1122 (1961).

Minimizing the free energy with respect to the strains,

$$\partial F/\partial \epsilon_i^{\mu,j}=0, \tag{12}$$

one finds

$$\epsilon_i^{\mu,j}=(1/c_j^{\mu})\textstyle\sum_l B_{j,l}^{\mu}\langle\mathcal{K}_i^{\mu,l}\rangle. \tag{13}$$

In Sec. 5 we shall show[11] that the statistical average of a TKO is proportional to a Kubic harmonic. That is

$$\langle\mathcal{K}_i^{\mu,l}\rangle=\langle\mathcal{Y}_{l(\zeta)}{}^0\rangle K_i^{\mu,l}(\zeta), \tag{14}$$

where $\mathcal{Y}_{l(\zeta)}{}^0$ is the spherical tensor operator which transforms identically to the Legendre polynomial P_l^0, with the magnetization direction (or the common axis of the sublattice magnetization) as the polar axis, as indicated by the subscript ζ in the average; $\langle\mathcal{Y}_{l(\zeta)}{}^0\rangle$ is a function of the temperature only. $K_i^{\mu,l}$ is the Kubic harmonic [a function of the angles θ and ϕ of the (sublattice) magnetization axis relative to axes fixed in the crystal] which transforms under the cubic group in the same way as the TKO. For compactness let

$$B_{j,l}^{\mu}\langle\mathcal{Y}_{l(\zeta)}{}^0\rangle\equiv\bar{B}_{j,l}^{\mu}(T). \tag{15}$$

Then

$$\epsilon_i^{\mu,j}=\frac{1}{c_j^{\mu}}\sum_l B_{j,l}^{\mu}K_i^{\mu,l}. \tag{16}$$

This is the formal solution for the strain components as a function of temperature and (sublattice) magnetization direction. In a later section we shall discuss this solution in greater detail, elaborating on the temperature dependence and considering some of the strains explicitly. For now, we obtain the general form of the effective spin Hamiltonian and of the free energy and specific heat, which result from substituting this solution for the strains into the Hamiltonian and the free energy.

First we find the self-consistent spin Hamiltonian. Substituting the strain, Eq. (16), into the general Hamiltonian, Eq. (10), we obtain

$$\begin{aligned}H=H_m+\tfrac{1}{2}\sum_{\mu}\sum_{j}\frac{1}{c_j^{\mu}}\sum_{l,l'}B_{j,l}^{\mu}B_{j,l'}^{\mu}\sum_i K_i^{\mu,l}K_i^{\mu,l'}\\ -\sum_{\mu}\sum_{j}\frac{1}{c_j^{\mu}}\sum_{l,l'}B_{j,l}^{\mu}\bar{B}_{j,l'}^{\mu}\sum_i K_i^{\mu,l'}\mathcal{K}_i^{\mu,l}\\ +\sum\kappa_l\mathcal{K}^{\alpha,l}.\end{aligned} \tag{17}$$

Let

$$\sum_i K_i^{\mu,l_1}K_i^{\mu,l_2}=\sum_l k_{l_1,l_2}^{\mu,l}K^{\alpha,l}. \tag{18}$$

We will give explicit expressions for the expansion coefficients $k_{l_1,l_2}^{\mu,l}$ in Sec. 4. Then

$$\begin{aligned}H=H_m+\tfrac{1}{2}\sum_{\mu}\sum_{j}\frac{1}{c_j^{\mu}}\sum_{l_1,l_2}\bar{B}_{j,l_1}^{\mu}\bar{B}_{j,l_2}^{\mu}\sum_l k_{l_1,l_2}^{\mu,l}K^{\alpha,l}\\ -\sum_{\mu}\sum_{j}\frac{1}{c_j^{\mu}}\sum_{l_1,l_2}\bar{B}_{j,l_1}^{\mu}\bar{B}_{j,l_2}^{\mu}\sum_i K_i^{\mu,l_2}\mathcal{K}_i^{\mu,l_1}\\ +\sum_l\kappa_l\mathcal{K}^{\alpha,l}.\end{aligned} \tag{19}$$

This is the self-consistent Hamiltonian for the spin operators. The free energy can be found either from it, or from Eq. (11). Substituting the strains into Eq. (11) we find

$$\begin{aligned}F=F_m-\tfrac{1}{2}\sum_{\mu}\sum_{j}\frac{1}{c_j^{\mu}}\sum_{l_1,l_2}\bar{B}_{j,l_1}^{\mu}\bar{B}_{j,l_2}^{\mu}\sum_l k_{l_1,l_2}^{\mu,l}K^{\alpha,l}\\ +\sum_l\bar{\kappa}_lK^{\alpha,l}.\end{aligned} \tag{20}$$

If we define the effective magnetocrystalline anisotropy coefficient $\bar{\kappa}_l^{\text{eff}}$ by

$$F=F_m+\sum_l\bar{\kappa}_l^{\text{eff}}K^{\alpha,l}, \tag{21a}$$

then

$$\bar{\kappa}_l^{\text{eff}}=\bar{\kappa}_l-\tfrac{1}{2}\sum_{\mu}\sum_{j}\frac{1}{c_j^{\mu}}\sum_{l_1,l_2}k_{l_1,l_2}^{\mu,l}B_{j,l_1}^{\mu}B_{j,l_2}^{\mu}. \tag{21b}$$

Whereas $\bar{\kappa}_l$ satisfies the famous $l(l+1)/2$ power law for the magnetocrystalline anisotropy[12] we see that the magnetoelastic coupling contributes additional terms of different temperature dependence, as we shall examine subsequently.

The physical source of this alteration in the temperature dependence of the anisotropy is as follows. For a given direction of the magnetization the crystal distorts under the influence of the magnetostrictive coupling, so that the symmetry is lower than cubic. This lower symmetry determines the temperature dependence of the anisotropy. It should be noted, however, that the magnetostrictive distortion is cubically modulated as the magnetization vector is rotated. Consequently, although the magnitude and temperature dependence of the anisotropy are influenced by the distortion, the observed anisotropy retains its over-all cubic symmetry.

The pair of Eqs. (16) and (21) constitute a complete formal solution of the problem. Equation (16) defines the strain induced by the magnetoelastic coupling and characterizes the change in crystal symmetry below the Curie temperature. Equations (21) for the free energy completely determine the thermodynamics of the system; thus $\bar{\kappa}_l^{\text{eff}}(T)$ is the effective anisotropy constant, with an altered temperature dependence, and the specific heat, which depends on the direction of the

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[11] See J. H. Van Vleck, Colloque International de Magnetisme de Grenoble, 1958 (unpublished).

[12] N. Akulov, Z. Physik 100, 197 (1936).

TABLE II. Table of spherical tensor operators, normalized to unity, from $l=0$ through $l=6$. The symbol $P\{\ \}$ means the sum of all permutations of the operators in the bracket, taken in first order. Thus $P\{S^+S_z^2\} \equiv S^+S_z^2+S_zS^+S_z+S_z^2S^+$.

$l=0$	$\mathcal{Y}_0^0=n_0^0 1$
$l=1$	$\mathcal{Y}_1^1=n_1^1 S^+$
	$\mathcal{Y}_1^0=n_1^1 S_z$
$l=2$	$\mathcal{Y}_2^2=n_2^2(S^+)^2$
	$\mathcal{Y}_2^1=\frac{n_2^2}{\sqrt{2}}P\{S^+S_z\}$
	$\mathcal{Y}_2^0=\frac{n_2^2}{\sqrt{6}}[P\{S^+S^-\}+2S_z^2]$
$l=3$	$\mathcal{Y}_3^3=n_3^3(S^+)^3$
	$\mathcal{Y}_3^2=\frac{n_3^3}{\sqrt{3}}P\{(S^+)^2S_z\}$
	$\mathcal{Y}_3^1=\frac{n_3^3}{(15)^{1/2}}[P\{(S^+)^2S^-\}+2P\{S^+S_z^2\}]$
	$\mathcal{Y}_3^0=\frac{n_3^3}{(10)^{1/2}}[P\{S^+S_zS^-\}+2S_z^3]$
$l=4$	$\mathcal{Y}_4^4=n_4^4(S^+)^4$
	$\mathcal{Y}_4^3=\frac{n_4^4}{2}P\{(S^+)^3S_z\}$
	$\mathcal{Y}_4^2=\frac{n_4^4}{2\times 7^{1/2}}[P\{(S^+)^3S^-\}+2P\{(S^+)^2S_z^2\}]$
	$\mathcal{Y}_4^1=\frac{n_4^4}{2\times 7^{1/2}}[P\{(S^+)^2S_zS^-\}+2P\{S^+S_z^3\}]$
	$\mathcal{Y}_4^0=\frac{n_4^4}{(70)^{1/2}}[P\{(S^+)^2(S^-)^2\}+2P\{S^+S_z^2S^-\}+4S_z^4]$
$l=5$	$\mathcal{Y}_5^5=n_5^5(S^+)^5$
	$\mathcal{Y}_5^4=\frac{n_5^5}{5^{1/2}}P\{(S^+)^4S_z\}$
	$\mathcal{Y}_5^3=\frac{n_5^5}{3(5)^{1/2}}[P\{(S^+)^4S^-\}+2P\{(S^+)^3S_z^2\}]$
	$\mathcal{Y}_5^2=\frac{n_5^5}{2(15)^{1/2}}[P\{(S^+)^3S_zS^-\}+2P\{(S^+)^2S_z^3\}]$
	$\mathcal{Y}_5^1=\frac{n_5^5}{(2\times 3\times 5\times 7)^{1/2}}[P\{(S^+)^3(S^-)^2\}+2P\{(S^+)^2S_z^2S^-\}+4P\{S^+S_z^4\}]$
	$\mathcal{Y}_5^0=\frac{n_5^5}{3(14)^{1/2}}[P\{(S^+)^2S_z(S^-)^2\}+2P\{S^+S_z^3S^-\}+4S_z^5]$
$l=6$	$\mathcal{Y}_6^6=n_6^6(S^+)^6$
	$\mathcal{Y}_6^5=\frac{n_6^6}{6^{1/2}}P\{(S^+)^5S_z\}$
	$\mathcal{Y}_6^4=\frac{n_6^6}{(6\times 11)^{1,2}}[P\{(S^+)^5S^-\}+2P\{(S^+)^4S_z^2\}]$
	$\mathcal{Y}_6^3=\frac{n_6^6}{(2\times 5\times 11)^{1/2}}[P\{(S^+)^4S_zS^-\}+2P\{(S^+)^3S_z^3\}]$
	$\mathcal{Y}_6^2=\frac{n_6^6}{3(5\times 11)^{1/2}}[P\{(S^+)^4(S^-)^2\}+2P\{(S^+)^3S_z^2S^-\}+4P\{(S^+)^2S_z^4\}]$
	$\mathcal{Y}_6^1=\frac{n_6^6}{6(11)^{1/2}}[P\{(S^+)^3S_z(S^-)^2\}+2P\{(S^+)^2S_z^3S^-\}+4P\{S^+S_z^5\}]$
	$\mathcal{Y}_6^0=\frac{n_6^6}{2(3\times 7\times 11)^{1/2}}[P\{(S^+)^3(S^-)^3\}+2P\{(S^+)^2S_z^2(S^-)^2\}+4P\{S^+S_z^4S^-\}+8S_z^6]$

magnetization, is of magnitude

$$c_V=c_{VM}-T\sum_l \frac{\partial^2 \bar{\kappa}_l^{\mathrm{eff}}(T)}{\partial T^2}K^{\alpha,l}. \tag{22}$$

In order to analyze the formal solution in detail we now proceed to study the TKO's explicitly, to obtain definite expressions for the expansion coefficients $k_{l_1,l_2}^{\mu,l}$, and to study the temperature dependence of the $\bar{B}_{j,l}^{\mu}$.

4. TENSOR KUBIC OPERATORS

The spherical harmonics $\mathcal{Y}_l^m(\theta,\phi)$ of given l form a set of basis functions for the $(2l+1)$ dimensional irreducible representation of the full (spherical) rotation group. Similarly, the spherical tensor operators $\mathcal{Y}_l^m$ of given l form an equivalent basis, standing in one-to-one correspondence with the spherical harmonics. The spherical tensor operator $\mathcal{Y}_l^m$ is a polynomial in the spin operators[13] S_x, S_y, S_z (or S^+, S_z, S^-). Although the rotation properties of the $\mathcal{Y}_l^m$ are identical to the $\mathcal{Y}_l^m$, and although it is only these rotation properties which enter into the analysis, the specific forms of the $\mathcal{Y}_l^m$ are sometimes useful in explicit calculations. We give the $\mathcal{Y}_l^m$ through $l=6$ in Table II.

The most convenient phase and normalization of the

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[13] In the application made here, the spin operators are those of a single representative ion in the crystal.

Table III. Tensor Kubic Operators expanded in spherical tensors.

Γ_α	$\mathcal{K}_1^{\alpha,4}=(7/12)^{1/2}\mathcal{Y}_4^0+(5/24)^{1/2}(\mathcal{Y}_4^4+\mathcal{Y}_4^{-4})$
	$\mathcal{K}_1^{\alpha,6}=(1/8)^{1/2}\mathcal{Y}_6^0-(7/16)^{1/2}(\mathcal{Y}_6^4+\mathcal{Y}_6^{-4})$
$\Gamma_{\beta'}$	$\mathcal{K}_1^{\beta',6}=(11/32)^{1/2}(\mathcal{Y}_6^2+\mathcal{Y}_6^{-2})-(5/32)^{1/2}(\mathcal{Y}_6^6+\mathcal{Y}_6^{-6})$
Γ_γ	$\mathcal{K}_1^{\gamma,2}=\mathcal{Y}_2^0$
	$\mathcal{K}_2^{\gamma,2}=(1/\sqrt{2})(\mathcal{Y}_2^2+\mathcal{Y}_2^{-2})$
	$\mathcal{K}_1^{\gamma,4}=(1/\sqrt{2})(\mathcal{Y}_4^2+\mathcal{Y}_4^{-2})$
	$\mathcal{K}_2^{\gamma,4}=(5/12)^{1/2}\mathcal{Y}_4^0-(7/24)^{1/2}(\mathcal{Y}_4^4+\mathcal{Y}_4^{-4})$
	$\mathcal{K}_1^{\gamma,6}=(7/8)^{1/2}\mathcal{Y}_6^0+\frac{1}{4}(\mathcal{Y}_6^4+\mathcal{Y}_6^{-4})$
	$\mathcal{K}_2^{\gamma,6}=(5/32)^{1/2}(\mathcal{Y}_6^2+\mathcal{Y}_6^{-2})+(11/32)^{1/2}(\mathcal{Y}_6^6+\mathcal{Y}_6^{-6})$
Γ_ϵ	$\mathcal{K}_1^{\epsilon,2}=+(i/\sqrt{2})(\mathcal{Y}_2^1+\mathcal{Y}_2^{-1})$
	$\mathcal{K}_2^{\epsilon,2}=-(1/\sqrt{2})(\mathcal{Y}_2^1-\mathcal{Y}_2^{-1})$
	$\mathcal{K}_3^{\epsilon,2}=-(i/\sqrt{2})(\mathcal{Y}_2^2-\mathcal{Y}_2^{-2})$
	$\mathcal{K}_1^{\epsilon,4}=+(i/4)(\mathcal{Y}_4^1+\mathcal{Y}_4^{-1})-i(7/16)^{1/2}(\mathcal{Y}_4^3+\mathcal{Y}_4^{-3})$
	$\mathcal{K}_2^{\epsilon,4}=(-1/4)(\mathcal{Y}_4^1-\mathcal{Y}_4^{-1})-(7/16)^{1/2}(\mathcal{Y}_4^3-\mathcal{Y}_4^{-3})$
	$\mathcal{K}_3^{\epsilon,4}=(-i/\sqrt{2})(\mathcal{Y}_4^2-\mathcal{Y}_4^{-2})$
	$\mathcal{K}_1^{\epsilon,6}=+i(5/256)^{1/2}(\mathcal{Y}_6^1+\mathcal{Y}_6^{-1})-i(9\sqrt{2}/32)(\mathcal{Y}_6^3+\mathcal{Y}_6^{-3})$ $+i[(2\times3\times5\times11)^{1/2}/32](\mathcal{Y}_6^5+\mathcal{Y}_6^{-5})$
	$\mathcal{K}_2^{\epsilon,6}=(-5^{1/2}/16)(\mathcal{Y}_6^1-\mathcal{Y}_6^{-1})-(9\sqrt{2}/32)(\mathcal{Y}_6^3-\mathcal{Y}_6^{-3})$ $-[(2\times3\times5\times11)^{1/2}/32](\mathcal{Y}_6^5-\mathcal{Y}_6^{-5})$
	$\mathcal{K}_3^{\epsilon,6}=(-i/\sqrt{2})(\mathcal{Y}_6^2-\mathcal{Y}_6^{-2})$
	$\mathcal{K}_1^{\epsilon,6'}=+i[(9\times11)^{1/2}/16](\mathcal{Y}_6^1+\mathcal{Y}_6^{-1})$ $+i[(2\times5\times11)^{1/2}/32](\mathcal{Y}_6^3+\mathcal{Y}_6^{-3})$ $+i[(2\times3)^{1/2}/32](\mathcal{Y}_6^5+\mathcal{Y}_6^{-5})$
	$\mathcal{K}_2^{\epsilon,6'}=-[(9\times11)^{1,2}/16](\mathcal{Y}_6^1-\mathcal{Y}_6^{-1})$ $+[(2\times5\times11)^{1/2}/32](\mathcal{Y}_6^3-\mathcal{Y}_6^{-3})$ $-[(2\times3)^{1/2}/32](\mathcal{Y}_6^5-\mathcal{Y}_6^{-5})$
	$\mathcal{K}_3^{\epsilon,6'}=-(i/\sqrt{2})(\mathcal{Y}_6^6-\mathcal{Y}_6^{-6})$
$\Gamma_{\delta'}$	$\mathcal{K}_1^{\delta',4}=+i(7^{1/2}/4)(\mathcal{Y}_4^1+\mathcal{Y}_4^{-1})+(i/4)(\mathcal{Y}_4^3+\mathcal{Y}_4^{-3})$
	$\mathcal{K}_2^{\delta',4}=(-7^{1/2}/4)(\mathcal{Y}_4^1-\mathcal{Y}_4^{-1})+\frac{1}{4}(\mathcal{Y}_4^3-\mathcal{Y}_4^{-3})$
	$\mathcal{K}_3^{\delta',4}=(-i/\sqrt{2})(\mathcal{Y}_4^4-\mathcal{Y}_4^{-4})$
	$\mathcal{K}_1^{\delta',6}=+i(3/32)^{1/2}(\mathcal{Y}_6^1+\mathcal{Y}_6^{-1})-i(15^{1/2}/8)(\mathcal{Y}_6^3+\mathcal{Y}_6^{-3})$ $-i(11^{1/2}/8)(\mathcal{Y}_6^5+\mathcal{Y}_6^{-5})$
	$\mathcal{K}_2^{\delta',6}=-(3/32)^{1/2}(\mathcal{Y}_6^1-\mathcal{Y}_6^{-1})-(15^{1/2}/8)(\mathcal{Y}_6^3-\mathcal{Y}_6^{-3})$ $+(11^{1/2}/8)(\mathcal{Y}_6^5-\mathcal{Y}_6^{-5})$
	$\mathcal{K}_3^{\delta',6}=-(i/\sqrt{2})(\mathcal{Y}_6^4-\mathcal{Y}_6^{-4})$

spherical tensors seems to be achieved by letting

$$S^+=-(S_x+iS_y)/\sqrt{2}, \tag{23a}$$

$$S^-=(S_x-iS_y)/\sqrt{2}. \tag{23b}$$

With this convention,

$$(S^-,S^+)=S_z, \tag{24a}$$

$$(S^+,S_z)=-S^+, \tag{24b}$$

$$(S^-,S_z)=S^-. \tag{24c}$$

Then[14] the highest order spherical tensor of a given degree is

$$\mathcal{Y}_l^l=n_l^l(S^+)^l. \tag{25}$$

The lower order tensors are found by successive application of the formula

$$\mathcal{Y}_l^{m-1}=\left(\frac{l(l+1)-m(m-1)}{2}\right)^{-1/2}(S^-,\mathcal{Y}_l^m). \tag{26}$$

Meckler[14] gives the normalization of the highest order tensor as

$$(n_l^l)^2=\frac{2^l(2l+1)!(2S-l)!}{(l!)^2(2S+l+1)!}. \tag{27}$$

With this normalization,

$$\mathrm{Tr}(\mathcal{Y}_l^m)^\dagger\mathcal{Y}_l^{m'}=\delta_{m,m'}. \tag{28}$$

The symbol † signifies the adjoint. It will be seen from the normalization formula that the operators $\mathcal{Y}_l^m$ are only supported by a spin of sufficient order that

$$2S\geqslant l, \tag{29}$$

and the same statement, of course, applies to the TKO's. In Table II we list the spherical tensor operators from $\mathcal{Y}_l^l$ down to $\mathcal{Y}_l^0$. Operators of negative order are found by the formula

$$\mathcal{Y}_l^{-m}=(-1)^m(\mathcal{Y}_l^m)^\dagger. \tag{30}$$

$\mathcal{Y}_l^{-m}$ is found from $\mathcal{Y}_l^m$ by simply interchanging S^+ and S^- in all formulas. It will be seen from the table that, if S_x is replaced by x, S_y by y, and S_z by z, the $\mathcal{Y}_l^m$ reduce to the classical Y_l^m, apart from normalization, through $\mathcal{Y}_4^3$. $\mathcal{Y}_4^2$ differs, and thereafter there are frequent departures. All $\mathcal{Y}_l^l$ reduce to Y_l^l, however.

The Kubic harmonics $K_i^{\mu,l}(\theta,\phi)\equiv K_i^{\mu,l}(\zeta)$ are linear combinations of the $Y_l^m(\theta,\phi)$ which form basis functions for the Γ_μ irreducible representation of the cubic group:

$$K_i^{\mu,l}(\zeta)=\sum_m a_{i,m}^{\mu,l}Y_l^m. \tag{31}$$

The expansion coefficients have been calculated by Bethe[15] and by Ebina and Tsuya.[16] Our phase convention is, however, slightly different from Ebina and Tsuya. We prefer to give the Y_l^m the same phase as the $\mathcal{Y}_l^m$. Thus, our Y_l^m differ from those of Ebina and Tsuya, and of Bethe, by the factor $(-1)^m$. Furthermore, in the three-dimensional representations our subscripts $i=1, 2, 3$ stand for x, y, z while in Ebina and Tsuya $i=1, 2, 3$ represent z, x, y, respectively.

The tensor Kubic operators $\mathcal{K}_i^{\mu,l}$ are linear combinations of the spherical tensor operators $\mathcal{Y}_l^m$ *with the same expansion coefficients*:

$$\mathcal{K}_i^{\mu,l}=\sum_m a_{i,m}^{\mu,l}\mathcal{Y}_l^m. \tag{32}$$

The $\mathcal{K}_i^{\mu,l}$ then stand in one-to-one correspondence with the $K_i^{\mu,l}$, and form an equivalent basis for Γ_μ. The explicit expansions of Eq. (32) are given in Table III.

In taking the direct product of the Kubic harmonics of Γ_μ with themselves, and extracting the fully sym-

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[14] A. Meckler, Suppl. Nuovo Cimento **12**, 1 (1959).

[15] H. A. Bethe, Ann. Physik **3**, 133 (1929).

[16] Y. Ebina and N. Tsuya, Repts. Research Inst. Elec. Commun, Tohoku Univ. **12**, 1 (1960).

metric combination, we are led to the quantity (see Eq. 18)

$$\sum_i K_i^{\mu,l_1} K_i^{\mu,l_2} = \sum_l k_{l_1,l_2}^{\mu,l} K^{\alpha,l}. \tag{33}$$

The equation above constitutes a statement that the summation on the left is fully symmetric, and that it, therefore, is a sum of Kubic harmonics belonging to Γ_α. We proceed to calculate the expansion coefficients by inserting Eq. (31) into Eq. (33):

$$\sum_{m_1,m_2} \sum_i a_{i,m_1}^{\mu,l_1} a_{i,m_2}^{\mu,l_2} Y_{l_1}^{m_1} Y_{l_2}^{m_2} = \sum_l k_{l_1,l_2}^{\mu,l} \sum_m a_{1,m}^{\alpha,l} Y_l^m. \tag{34}$$

However, the addition theorem for the spherical harmonics is[17]

$$Y_{l_1}^{m_1} Y_{l_2}^{m_2} = \sum_{l,m} \left(\frac{(2l_1+1)(2l_2+1)}{4\pi(2l+1)} \right)^{1/2} \langle l_1 l_2 0 0 | l_1 l_2 l 0 \rangle \times \langle l_1 l_2 m_1 m_2 | l_1 l_2 l 0 \rangle Y_l^m, \tag{35}$$

where $\langle \cdots | \cdots \rangle$ indicates a Clebsch-Gordan coefficient. Hence,

$$\sum_l \left(\frac{(2l_1+1)(2l_2+1)}{4\pi(2l+1)} \right)^{1/2} \langle l_1 l_2 0 0 | l_1 l_2 l 0 \rangle \times \sum_m \sum_{m_1,m_2} \sum_i a_{i,m_1}^{\mu,l_1} a_{i,m_2}^{\mu,l_2} \langle l_1 l_2 m_1 m_2 | l_1 l_2 l m \rangle \mathcal{Y}_l^m = \sum_l k_{l_1,l_2}^{\mu,l} \sum_m a_{1,m}^{\alpha,l} \mathcal{Y}_l^m. \tag{36}$$

Because of the orthogonality of the Y_l^m we can equate coefficients of Y_l^0 to obtain

$$k_{l_1,l_2}^{\mu,l} = \left(\frac{(2l_1+1)(2l_2+1)}{4\pi(2l+1)} \right)^{1/2} \langle l_1 l_2 0 0 | l_1 l_2 l 0 \rangle \times \sum_m \sum_i \frac{a_{i,m}^{\mu,l_1} a_{i,-m}^{\mu,l_2}}{a_{1,0}^{\alpha,l}} \langle l_1 l_2 m -m | l_1 l_2 l 0 \rangle. \tag{37}$$

An alternate expression, sometimes more convenient, is obtained by equating coefficients of Y_l^4 in Eq. (36):

$$k_{l_1,l_2}^{\mu,l} = \left(\frac{(2l_1+1)(2l_2+1)}{4\pi(2l+1)} \right)^{1/2} \frac{\langle l_1 l_2 0 0 | l_1 l_2 l 0 \rangle}{a_{1,4}^{\alpha,l}} \times \sum_m \langle l_1 l_2 m (4-m) | l_1 l_2 l 4 \rangle \sum_i a_{i,m}^{\mu,l_1} a_{i,4-m}^{\mu,l_2}. \tag{38}$$

The Clebsch-Gordan coefficients which occur in Eqs. (37) and (38) can be evaluated by use of the usual expressions, as given by Edmonds[18] or Rose[17] or can be found in the tables by Shimpuku[19] and by Rotenberg, Bevins, Metropolis, and Wooten.[20] Some of the terms can be seen to vanish, and others can be simplified. In particular, the isotropic terms, which will add to the specific heat, are much reduced by the use of the symmetry relations and special values of the Clebsch-Gordan coefficients.[21,22] Thus, by means of the symmetry relation

$$\langle l_1 l_2 m_1 m_2 | l_1 l_2 l m \rangle = (2l+1/2l_2+1)^{1/2} (-1)^{l_1-m_1} \times \langle l_1 l m_1 -m | l_1 l l_2 -m_2 \rangle, \tag{39}$$

and the special value

$$\langle l_1 0 m_1 0 | l_1 0 l m \rangle = \delta_{l_1,l} \delta_{m_1,m}, \tag{40}$$

the isotropic coefficients become

$$k_{l_1,l_2}^{\mu,0} = \frac{1}{(4\pi)^{1/2}} \sum_m (-1)^m \sum_i a_{i,m}^{\mu,l_1} a_{i,-m}^{\mu,l_2} \delta_{l_1,l_2}. \tag{41}$$

Now the normalization of the Kubic harmonics requires that

$$\begin{aligned} 1 = \langle K_i^{\mu,l} | K_i^{\mu,l} \rangle &= \sum_{m,m'} a_{i,m}^{\mu,l*} a_{i,m'}^{\mu,l} \langle Y_l^m | Y_l^{m'} \rangle \\ &= \sum_m a_{i,m}^{\mu,l*} a_{i,m}^{\mu,l} \qquad (42) \\ 1 &= \sum_m (-1)^m a_{i,m}^{\mu,l} a_{i,-m}^{\mu,l}. \end{aligned}$$

Hence, if we let d_μ be the dimensionality of the μth irreducible representation,

$$k_{l_1,l_2}^{\mu,0} = [d_\mu / (4\pi)^{1/2}] \delta_{l_1,l_2}. \tag{43}$$

In addition, by means of the symmetry relations between the Clebsch-Gordan coefficients, it can be shown that

$$k_{l_1,l_2}^{\alpha,l} = k_{l_2,l_1}^{\alpha,l} = k_{l,,l}^{\alpha,l_2}, \quad l_1,\ l_2,\ l \neq 0. \tag{44}$$

In general, the expansion coefficients $k_{l_1,l_2}^{\mu,l}$ must be calculated by means of Eqs. (37) or (38), and are given in Table IV [actually, $(4\pi)^{1/2} k_{l_1,l_2}^{\mu,l}$]. These expressions for the coefficients $k_{l_1,l_2}^{\mu,l}$ can then be used to evaluate the free energy [Eq. (21a)], the effective anisotropy constants [Eq. (21b)] and the specific heat [Eq. (22)].

5. TEMPERATURE DEPENDENCE

In the equations for the strains, the free energy, and the specific heat there occur the quantities $\bar{B}_{j,l}^{\mu}(T)$, the temperature-dependent magnetoelastic coupling

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[17] M. E. Rose, *Elementary Theory of Angular Momentum* (John Wiley & Sons, Inc., New York, 1957), p. 61.

[18] A. R. Edmonds, *Angular Momentum in Quantum Mechanics* (Princeton University Press, Princeton, New Jersey, 1957), p. 45.

[19] T. Shimpuku, Suppl. Progr. Theoret. Phys. (Kyoto) **13**, 1 (1960).

[20] M. Rotenberg, R. Bevins, N. Metropolis, and J. K. Wooten, *The 3j* and *6j Symbols* (Massachusetts Institute of Technology Press, Cambridge, Massachusetts, 1959).

[21] G. Racah, Phys. Rev. **62**, 438 (1942).

[22] L. Eisenbud, Ph.D. thesis, Princeton University, 1948 (unpublished).

TABLE IV. Table of $(4\pi)^{1/2}k_{l_1,l_2}^{\mu,l}$, the expansion coefficients for sums (over the dimensions of the μth irreducible representation) of products of TKO's expanded in fully cubic TKO's. [See Eq. (18)].

	$(4\pi)^{1/2}k_{l_1,l_2}^{\alpha,l}$ $l_1=4,\ l_2=4$	$l_1=6,\ l_2=6$
$l=4$	$\frac{18(21)^{1/2}}{11\times 13}$	$\frac{-21(21)^{1/2}}{11\times 17}$
$l=6$	$\frac{20(2)^{1/2}}{11(13)^{1/2}}$	$\frac{-40(2\times 13)^{1/2}}{11\times 17\times 19}$

	$(4\pi)^{1/2}k_{l_1,l_2}^{\beta',l}$ $l_1=6,\ l_2=6$
$l=4$	$\frac{(21)^{1/2}}{17}$
$l=6$	$\frac{-5\times 8(2\times 13)^{1/2}}{17\times 19}$

		$(4\pi)^{1/2}k_{l_1,l_2}^{\gamma,4}$	
$l_1 \backslash l_2$	2	4	6
2	$2\left(\frac{3}{7}\right)^{1/2}$	0	$\frac{10}{11}\left(\frac{2\times 3\times 5}{13}\right)^{1/2}$
4		$\frac{4\times 9}{11\times 13}\left(\frac{3}{7}\right)^{1/2}$	0
6			$\frac{2\times 19}{11\times 17}(3\times 7)^{1/2}$

		$(4\pi)^{1/2}k_{l_1,l_2}^{\gamma,6}$	
$l_1 \backslash l_2$	2	4	6
2	0	0	$\frac{4}{11}\left(\frac{7}{5}\right)^{1/2}$
4		$-\frac{32}{11}\left(\frac{2}{13}\right)^{1/2}$	0
6			$\frac{5\times 48}{11\times 17\times 19}(2\times 13)^{1/2}$

			$(4\pi)^{1/2}k_{l_1,l_2}^{\epsilon\,4}$	
$l_1 \backslash l_2$	2	4	6	6′
2	$-2\left(\frac{3}{7}\right)^{1/2}$	$\frac{-30}{11(7)^{1/2}}$	$\frac{5}{2\times 11}\left(\frac{3\times 13}{2}\right)^{1/2}$	$\frac{15}{2}\left(\frac{15}{2\times 11\times 13}\right)^{1/2}$
4		$\frac{-27}{11}\left(\frac{3}{7}\right)^{1/2}$	$\frac{15}{11}\left(\frac{2}{13}\right)^{1/2}$	$-3\left(\frac{10}{11\times 13}\right)^{1/2}$
6			$\frac{-59}{2\times 11\times 17}(3\times 7)^{1/2}$	$\frac{3}{2\times 17}\left(\frac{3\times 5\times 7}{11}\right)^{1/2}$
6′				$\frac{9}{2\times 17}(21)^{1/2}$

			$(4\pi)^{1/2}k_{l_1,l_2}^{\epsilon,6}$	
$l_1 \backslash l_2$	2	4	6	6′
2	0	$\frac{30}{11}\left(\frac{6}{13}\right)^{1/2}$	$\frac{-15(7)^{1/2}}{2\times 11}$	$\frac{3}{2}\left(\frac{7}{5\times 11}\right)^{1/2}$
4		$\frac{15}{11}\left(\frac{2}{13}\right)^{1/2}$	$\frac{3(21)^{1/2}}{4\times 11\times 17}$	$\frac{-27}{4\times 17}\left(\frac{3\times 5\times 7}{11}\right)^{1/2}$
6			$\frac{3\times 5\times 31}{11\times 17\times 19}(2\times 13)^{1/2}$	$\frac{3\times 5\times 7}{17\times 19}\left(\frac{2\times 5\times 13}{11}\right)^{1/2}$
6′				$\frac{-3\times 5}{17\times 19}(2\times 13)^{1/2}$

		$(4\pi)^{1/2}k_{l_1,l_2}^{\delta',l}$	
	$l_1=4\ \ l_2=4$	$l_1=4\ \ l_2=6$	$l_1=6\ \ l_2=6$
$l=4$	$\frac{3\times 9}{11\times 13}(3\times 7)^{1/2}$	$\frac{-2\times 3}{11}\left(\frac{3\times 5\times 7}{13}\right)^{1/2}$	$\frac{-3\times 16}{11\times 17}(3\times 7)^{1/2}$
$l=6$	$\frac{-3}{2\times 11(13)^{1/2}}$	$\frac{7\times 9}{11\times 17}\left(\frac{5}{2}\right)^{1/2}$	$\frac{-3\times 4\times 5}{11\times 17\times 19}(2\times 13)^{1/2}$

coefficients. The temperature dependence arises in taking the average value of a TKO in the unperturbed density matrix. The important feature of the unperturbed density matrix is that it describes a system with the average value of the (sublattice) magnetization along some axis ζ, and that it has azimuthal symmetry around this axis.

Expressing the TKO in spherical tensor operators,

$$\langle \mathcal{K}_i^{\mu,l}\rangle = \sum_m a_{i,m}^{\mu,l}\langle \mathcal{Y}_l^m\rangle. \tag{45}$$

The spherical tensor operator $\mathcal{Y}_l^m$ can now be expressed in a new coordinate system with polar axis along ζ. Let $T_{l(\zeta)}^{m'}$ be the spherical tensor operators in this new coordinate system. Then

$$\begin{aligned}\mathcal{Y}_l^m &= \sum_{m'}\langle \mathcal{Y}_{l(\zeta)}^{m'} | \mathcal{Y}_l^m\rangle \mathcal{Y}_{l(\zeta)}^{m'} \\ &= \sum_{m'}\langle Y_{l(\zeta)}^{m'} | Y_l^m\rangle \mathcal{Y}_{l(\zeta)}^{m'},\end{aligned} \tag{46}$$

where the expansion coefficient $\langle \mathcal{Y}_{l(\zeta)}^{m'} | \mathcal{Y}_l^m\rangle$ is identical to the corresponding expansion coefficient for the spherical harmonics, and can, therefore, be written as the scalar product of two spherical harmonics $\langle Y_{l(\zeta)}^{m'} | Y_l^m\rangle$. Taking the average value of $\mathcal{Y}_l^m$ with respect to the unperturbed density matrix

$$\langle \mathcal{Y}_l^m\rangle = \sum_{m'}\langle Y_{l(\zeta)}^{m'} | Y_l^m\rangle\langle \mathcal{Y}_{l(\zeta)}^{m'}\rangle. \tag{47}$$

However, $\mathcal{Y}_{l(\zeta)}^{m'}$ transforms as $e^{im'\phi}$ under a rotation of ϕ around the ζ axis, whence $\langle \mathcal{Y}_{l(\zeta)}^{m'}\rangle = 0$ unless $m'=0$. Furthermore,[10]

$$\langle Y_{l(\zeta)}^0 | Y_l^m\rangle = Y_l^m(\zeta), \tag{48}$$

where $Y_l^m(\zeta)$ is the spherical harmonic of the angles θ, ϕ of the (sublattice) magnetization axis relative to the axes fixed in the cubic crystal. Hence,

$$\langle \mathcal{K}_i^{\mu,l}\rangle = \langle \mathcal{Y}_{l(\zeta)}^0\rangle \sum_m a_{i,m}^{\mu,l} Y_l^m(\zeta), \tag{49}$$

or

$$\langle \mathcal{K}_i^{\mu,l}\rangle = K_i^{\mu,l}(\zeta)\langle \mathcal{Y}_{l(\zeta)}^0\rangle. \tag{50}$$

This expression is the direct analog of a similar classical relation derived by Van Vleck,[11] and, as he has shown, it is the basis of the tenth-power law for the lowest order anisotropy coefficient. In fact, the temperature dependence of $\langle \mathcal{Y}_{l(\zeta)}^0\rangle$ is the same as that of $M^{l(l+1)/2}$ at very low temperatures, where M is the (sublattice) magnetization. Thus, we find from Table II that $\mathcal{Y}_{2(\zeta)}^0 \sim 3S_\zeta^2 - S(S+1)$. In terms of the spin deviation operator σ, $S_\zeta = S-\sigma$, and only the two states $\sigma = 0, 1$ are important at very low temperatures. Hence $S_\zeta^2 = S^2(1-\sigma)+(S-1)^2\sigma$ and

$$\langle \mathcal{Y}_{2(\zeta)}^0\rangle \sim (2S-1)S[1-3\langle\sigma\rangle/S] \simeq (M/M_0)^3.$$

At sufficiently low temperatures, the temperature dependences of all the magnetoelastic coupling coefficients, which are related to $\langle \mathcal{Y}_{l(\mathfrak{r})}{}^0\rangle$ by Eq. (15), are determined by this power law. In summary, if the first excited state in the space of two neighboring spins maintains the two spins parallel, then

$$\frac{B_{j,l}{}^{\mu}(T)}{\bar{B}_{j,l}{}^{\mu}(0)}=\left(\frac{M(T)}{M(0)}\right)^{l(l+1)/2}. \tag{51}$$

This relation constitutes the proof, as shown by Kittel and Van Vleck,[23] that the magnetostriction coefficient, like the anisotropy coefficient $\bar{\kappa}_l$, should follow the $l(l+1)/2$ power law.[24]

Though the proof of Eq. (51) is rigorously valid only for small perturbations, and at low temperatures, an exact calculation[25] for the case of a spin one Hamiltonian shows that, for the model employed, the $l(l+1)/2$ power law is fairly accurate even for relatively large perturbation, and is roughly followed almost up to the Curie temperature.

In a previous paper[26] we have also calculated the equivalent classical average, $\langle Y_{l(\mathfrak{r})}{}^0\rangle$, in the internal field approximation, and shown that

$$\langle Y_{l(\mathfrak{r})}{}^0\rangle=\frac{I_{l+\frac{1}{2}}[3(T_c/T)m_0]}{I_{1/2}[3(T_c/T)m_0]}\equiv\hat{I}_{(2l+1)/2}[3(T_c/T)m_0], \tag{52}$$

where I_l is the hyperbolic Bessel function, and m_0 is the reduced magnetization:

$$m_0\equiv M(T)/M(0). \tag{53}$$

This approximation, which should be rather good just below the Curie temperature, reduces to Eq. (51) at low temperatures. For the ferromagnet this equation can be applied directly, but for the antiferromagnet the argument of $\hat{I}_{(2l+1)/2}$ must be replaced by the molecular field as given by the Neél theory.

In order to compare theory with experiment, we examine the dependence of magnetostriction on magnetization rather than on temperature. The theoretical curve relates magnetostriction to temperature; it must be augmented with the corresponding theoretical dependence of magnetization on temperature (a Langevin function) so that the temperature can be eliminated parametrically. The resulting plot of $\hat{I}_{(2l+1)/2}$ as a function of the magnetization is given in Fig. 1, and the comparison with the data on nickel will be made in Sec. 7.

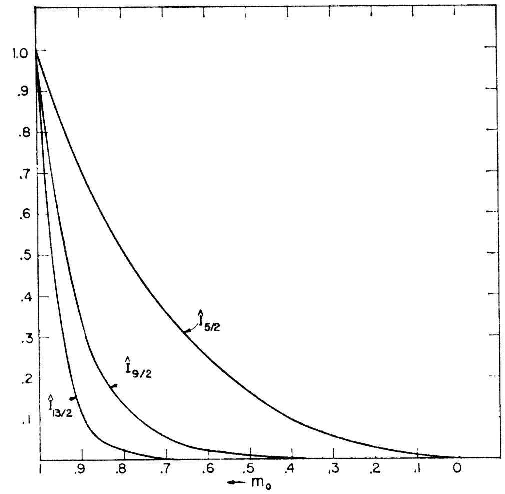

FIG. 1. The reduced hyperbolic Bessel functions $I_{(2l+1)/2}/I_{1/2}$ as a function of the reduced magnetization m_0. $l=2, 4, 6$.

The effective anisotropy coefficients Eq. (21) and the specific heat Eq. (22) involve sums over l_1 and l_2 consistent with a given l; consequently, although the individual $\bar{B}_{j,l_1}{}^{\mu}$ follow the $l(l+1)/2$ power law, the effective anisotropy coefficient and specific heat need not behave so simply. In fact, inserting the low-temperature approximation (Eq. 51) for the temperature dependence of the $\bar{B}_{j,l}{}^{\mu}$ into $\kappa_l{}^{\text{eff}}$ (from Eq. 21), and using the fact that in this temperature range $m_0{}^n$ is approximately equal to $1-n\delta m_0$, we find

$$\bar{\kappa}_l{}^{\text{eff}}=\bar{\kappa}_l{}^{\text{eff}}(0)\left\{1-\left[\frac{l(l+1)}{2}\left(\frac{\kappa_l}{\kappa_l{}^{\text{eff}}(0)}\right)\right.\right.$$
$$-\tfrac{1}{2}\sum_{\mu}\sum_{j}\sum_{l_1,l_2}\left(\frac{l_1(l_1+1)}{2}+\frac{l_2(l_2+1)}{2}\right)$$
$$\left.\left.\times\frac{k_{l_1,l_2}{}^{\mu,l}B_{j,l_1}{}^{\mu}B_{j,l_2}{}^{\mu}}{\bar{\kappa}_l{}^{\text{eff}}(0)c_j{}^{\mu}}\right]\delta m_0\right\}. \tag{54}$$

The quantity in the square bracket is the effective power of the (sublattice) magnetization.

The temperature dependence of the magnetoelastic contributions to the specific heat can also be found in the low-temperature region with the same generality. We have that

$$C_V=C_{VM}-T\sum_{l}\frac{\partial^2\bar{\kappa}_l{}^{\text{eff}}}{\partial T^2}K^{\alpha,l}. \tag{55}$$

There are magnetoelastic contributions both to the isotropic and cubic specific heat, though κ_l contributes only to the anisotropic terms. If we let

$$C_V=C_{VM}+\sum_{l=0}C_V{}^lK^{\alpha,l}, \tag{56}$$

then

$$C_V{}^l=-T(\partial^2/\partial T^2)\bar{\kappa}_l{}^{\text{eff}}, \tag{57}$$

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[23] C. Kittel and J. H. Van Vleck, Phys. Rev. **118**, 1231 (1960).

[24] In the antiferromagnet the ground state is not one of antiparallel arrangement of spins, but contains zero-point fluctuations. P. Pincus [Phys. Rev. **113**, 769 (1959)] has shown that this replaces the denominators in Eq. (51) by the values in the fully antiparallel arrangement. But as this equation is then true at any temperature, Pincus shows that it can be evaluated at 0°K, and the properties of the antiferromagnetic ground state eliminated, thus removing the apparent complication of the antiferromagnetic ground state and restoring Eq. (51).

[25] E. R. Callen, J. Appl. Phys. **33**, 832 (1962).

[26] E. R. Callen and H. B. Callen, J. Phys. Chem. Solids **16**, 310 (1960).

where $\bar{\kappa}_l^{\text{eff}}$ is again given by (21b) or at low temperatures, by (54).

In the low-temperature region in which this power law is appropriate the magnetization of a ferromagnet varies with the temperature, according to spin-wave theory, as

$$m_0(T)=1-rT^{3/2}. \tag{58}$$

Hence, when spin-wave theory applies,

$$C_V{}^l=\tfrac{1}{2}\,\tfrac{3}{2}r\Bigg[\frac{l(l+1)}{2}\kappa_l-\tfrac{1}{2}\sum_\mu\sum_i\frac{1}{c_j{}^\mu}\sum_{l_1,l_2}k_{l_1,l_2}{}^{\mu,l}\times B_{j,l_1}{}^\mu B_{j,l_2}{}^\mu\left(\frac{l_1(l_1+1)}{2}+\frac{l_2(l_2+1)}{2}\right)\Bigg]T^{1/2}. \tag{59}$$

The coefficient r has been evaluated for the cubic lattices.[27]

For an antiferromagnet spin-wave theory replaces Eq. (58) by an exponential temperature dependence, whence $C_V{}^l$ also depends exponentially on the temperature.

6. EXTERNAL STRAINS AND MAGNETOSTRICTION

The external strains support only the representations α, γ, and ϵ, and are indicated by the modes numbered zero. In this case Eq. (16) becomes

$$\epsilon_i{}^{\mu,0}=\frac{1}{c_0{}^\mu}\sum_l\bar{B}_{0,l}{}^\mu K_i{}^{\mu,l},\quad \mu=\alpha,\,\gamma,\,\epsilon. \tag{60}$$

To convert to the conventional strains we recall that

$$\epsilon_1{}^{\alpha,0}=\text{trace}\,\epsilon=\delta V/V=\epsilon_{xx}+\epsilon_{yy}+\epsilon_{zz}, \tag{61a}$$

that

$$\epsilon_1{}^{\gamma,0}=5^{1/2}[\epsilon_{zz}-\tfrac{1}{2}(\epsilon_{xx}+\epsilon_{yy})],\qquad \epsilon_2{}^{\gamma,0}=\tfrac{1}{2}(15)^{1/2}[\epsilon_{xx}-\epsilon_{yy}], \tag{61b}$$

and that

$$\epsilon_1{}^{\epsilon,0}=\epsilon_{yz};\quad \epsilon_2{}^{\epsilon,0}=\epsilon_{zx};\quad \epsilon_3{}^{\epsilon,0}=\epsilon_{xy}. \tag{61c}$$

By these relations the external strains can all be found in terms of the elastic coefficients and the magnetoelastic coupling coefficients. They depend upon the temperature through Eq. (15) and upon the magnetization direction through the factors $K_i{}^{\mu,l}(\zeta)$. By means of Eqs. (61), one derives readily that

$$\epsilon_{xx}=\frac{1}{3}\frac{1}{c_0{}^\alpha}\sum_l B_{0,l}{}^\alpha K_1{}^{\alpha,l}-\frac{1}{(15)^{1/2}}\frac{1}{c_0{}^\gamma}\sum_l\bar{B}_{0,l}{}^\gamma\left(\frac{1}{\sqrt{3}}K_1{}^{\gamma,l}-K_2{}^{\gamma,l}\right), \tag{62a}$$

$$\epsilon_{yy}=\frac{1}{3}\frac{1}{c_0{}^\alpha}\sum_l\bar{B}_{0,l}{}^\alpha K_1{}^{\alpha,l}-\frac{1}{(15)^{1/2}}\frac{1}{c_0{}^\gamma}\sum_l\bar{B}_{0,l}{}^\gamma\left(\frac{1}{\sqrt{3}}K_1{}^{\gamma,l}+K_2{}^{\gamma,l}\right), \tag{62b}$$

$$\epsilon_{zz}=\frac{1}{3}\frac{1}{c_0{}^\alpha}\sum_l\bar{B}_{0,l}{}^\alpha K_1{}^{\alpha,l}+\frac{2}{3(5)^{1/2}}\frac{1}{c_0{}^\gamma}\sum_l\bar{B}_{0,}{}^{l\gamma}K_1{}^{\gamma,l}. \tag{62c}$$

The external shears, which are proper basis functions for the ϵ representation, are found directly by Eqs. (60) and (61c).

Letting ξ_i be the direction cosines of the measurement direction with respect to the cubic-crystal axes, the fractional change in length of the crystal at saturation is given by

$$\frac{\delta l}{l}=\sum_{i\geq j}\epsilon_{ij}\xi_i\xi_j \tag{63}$$

$$=\frac{1}{3}\frac{1}{c_0{}^\alpha}\sum_l\bar{B}_{0,l}{}^\alpha K_1{}^{\alpha,l}-\frac{1}{(15)^{1/2}}\frac{1}{c_0{}^\gamma}\sum_l\bar{B}_{0,l}{}^\gamma\left(\frac{1}{\sqrt{3}}K_1{}^{\gamma,l}-K_2{}^{\gamma,l}\right)\xi_1{}^2-\frac{1}{(15)^{1/2}}\frac{1}{c_0{}^\gamma}\sum_l\bar{B}_{0,l}{}^\gamma\left(\frac{1}{\sqrt{3}}K_1{}^{\gamma,l}+K_2{}^{\gamma,l}\right)\xi_2{}^2+\frac{2}{3(5)^{1/2}}\frac{1}{c_0{}^\gamma}\sum_l\bar{B}_{0,l}{}^\gamma K_1{}^{\gamma,l}\xi_3{}^2+\frac{1}{c_0{}^\epsilon}\sum_l\bar{B}_{0,l}{}^\epsilon(K_3{}^{\epsilon,l}\xi_1\xi_2+K_1{}^{\epsilon,l}\xi_2\xi_3+K_2{}^{\epsilon,l}\xi_3\xi_1). \tag{64}$$

The elastic constants $c_0{}^\mu$ are related to the conventional elastic constants as in Eq. (6). These results are similar to those of Becker and Döring,[1] Kittel,[28] Lee,[29] and Birss.[30]

The Kubic harmonics are convenient for theoretical analysis because they are orthonormal, relate the magnetostriction coefficients simply to the magnetoelastic coupling coefficients, and separate the various temperature dependences. However, they are not the polynomials in terms of which magnetostriction is usually expressed. For convenience, we now recall the conventional definitions and we give the explicit relationships between the two sets of polynomials. To avoid reference to a fiducial state of random alignment, we choose magnetostriction coefficients with no corrections

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[27] J. Van Kranendonk and J. H. Van Vleck, Rev. Mod. Phys. **30**, 1 (1958).

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

[29] E. W. Lee, in *Reports on Progress in Physics* (The Physical Society, London, 1955), Vol. 18, p. 184.

[30] R. R. Birss, in *Advances in Physics*, edited by N. F. Mott (Taylor and Francis Ltd., London, 1959), Vol. 8, p. 252.

for "average values." Let

$$
\begin{aligned}
\frac{\delta l}{l}=\Lambda_1\Bigg[&\left(-\frac{1}{3(5)^{1/2}}K_1{}^{\gamma,2}+\frac{1}{(15)^{1/2}}K_2{}^{\gamma,2}\right)\xi_1{}^2\\
&+\left(-\frac{1}{3(5)^{1/2}}K_1{}^{\gamma,2}-\frac{1}{(15)^{1/2}}K_2{}^{\gamma,2}\right)\xi_2{}^2+\frac{2}{3(5)^{1/2}}K_1{}^{\gamma,2}\xi_3{}^2\Bigg]\\
&+\Lambda_2[K_3{}^{\epsilon,2}\xi_1\xi_2+\text{c.p.}]+\tfrac{1}{3}\Lambda_3K_1{}^{\alpha,4}\\
&+\Lambda_4\Bigg[\left(-\frac{1}{3(5)^{1/2}}K_1{}^{\gamma,4}+\frac{1}{(15)^{1/2}}K_2{}^{\gamma,4}\right)\xi_1{}^2\\
&+\left(-\frac{1}{3(5)^{1/2}}K_1{}^{\gamma,4}-\frac{1}{(15)^{1/2}}K_2{}^{\gamma,4}\right)\xi_2{}^2+\frac{2}{3(5)^{1/2}}K_1{}^{\gamma,4}\xi_3{}^2\Bigg]\\
&+\Lambda_5[K_3{}^{\epsilon,4}\xi_1\xi_2+\text{c.p.}]+\tfrac{1}{3}\Lambda_6K_1{}^{\alpha,6}\\
&+\Lambda_7\Bigg[\left(-\frac{1}{3(5)^{1/2}}K_1{}^{\gamma,6}+\frac{1}{(15)^{1/2}}K_2{}^{\gamma,6}\right)\xi_1{}^2\\
&+\left(-\frac{1}{3(5)^{1/2}}K_1{}^{\gamma,6}-\frac{1}{(15)^{1/2}}K_2{}^{\gamma,6}\right)\xi_2{}^2+\frac{2}{3(5)^{1/2}}K_1{}^{\gamma,6}\xi_3{}^2\Bigg]\\
&+\Lambda_8[K_3{}^{\epsilon,6}\xi_1\xi_2+\text{c.p.}]+\Lambda_9[K_3{}^{\epsilon,6'}\xi_1\xi_2+\text{c.p.}]. \qquad (65)
\end{aligned}
$$

Then,

$$
\begin{aligned}
\Lambda_1&=(1/c_0{}^{\gamma})\bar{B}_{0,2}{}^{\gamma},\\
\Lambda_2&=(1/c_0{}^{\epsilon})\bar{B}_{0,2}{}^{\epsilon},\\
\Lambda_3&=(1/c_0{}^{\alpha})\bar{B}_{0,4}{}^{\alpha},\\
\Lambda_4&=(1/c_0{}^{\gamma})\bar{B}_{0,4}{}^{\gamma},\\
\Lambda_5&=(1/c_0{}^{\epsilon})\bar{B}_{0,4}{}^{\epsilon}, \qquad (66\text{a–i})\\
\Lambda_6&=(1/c_0{}^{\alpha})\bar{B}_{0,6}{}^{\alpha},\\
\Lambda_7&=(1/c_0{}^{\gamma})\bar{B}_{0,6}{}^{\gamma},\\
\Lambda_8&=(1/c_0{}^{\epsilon})\bar{B}_{0,6}{}^{\epsilon},\\
\Lambda_9&=(1/c_0{}^{\epsilon})\bar{B}_{0,6'}{}^{\epsilon}.
\end{aligned}
$$

On the other hand, the magnetostriction is usually developed in homogeneous polynomials in the direction cosines of the (sublattice) magnetization, ζ_i, as

$$
\begin{aligned}
\delta l/l=C_1\textstyle\sum_i\zeta_i{}^2\xi_i{}^2&+C_2(\zeta_1\zeta_2\xi_1\xi_2+\text{c.p.})\\
&+C_3(\zeta_1{}^2\zeta_2{}^2+\text{c.p.})+C_4\textstyle\sum_i\zeta_i{}^4\xi_i{}^2\\
&+C_5(\zeta_1\zeta_2\zeta_3{}^2\xi_1\xi_2+\text{c.p.})+C_6\zeta_1{}^2\zeta_2{}^2\zeta_3{}^2\\
&+C_7\textstyle\sum_i\zeta_i{}^6\xi_i{}^2+C_8(\zeta_1\zeta_2\zeta_3{}^4\xi_1\xi_2+\text{c.p.})\\
&+C_9(\zeta_1{}^3\zeta_2{}^3\xi_1\xi_2+\text{c.p.}). \qquad (67)
\end{aligned}
$$

This is the form given by Birss[30] and by Vautier.[31] The matrices which transform the C_i into Λ_j are, from Γ_α and Γ_γ,

$$
\left\{\begin{matrix}
1 & 0 & 2\times3/7 & 0 & 5/7\\
0 & -\dfrac{2\times3^{1/2}}{5\times7^{1/2}} & 0 & \dfrac{-2\times3^{1/2}}{5\times11(7)^{1/2}} & 0\\
0 & 0 & \dfrac{2}{7\times3^{1/2}} & 0 & \dfrac{2\times5\times3^{1/2}}{7\times11}\\
0 & 0 & 0 & \dfrac{4\times2^{1/2}}{7\times11(13)^{1/2}} & \dfrac{4\times2^{1/2}}{7\times11(13)^{1/2}}\\
0 & 0 & 0 & 0 & \dfrac{2(2\times5)^{1/2}}{11(7\times13)^{1/2}}
\end{matrix}\right\}
\begin{pmatrix}C_1\\C_3\\C_4\\C_6\\C_7\end{pmatrix}
=\frac{1}{(4\pi)^{1/2}}\begin{pmatrix}\Lambda_1\\\Lambda_3\\\Lambda_4\\\Lambda_6\\\Lambda_7\end{pmatrix}, \qquad (68)
$$

and from Γ_ϵ,

$$
\left\{\begin{matrix}
\dfrac{1}{(3\times5)^{1/2}} & \dfrac{1}{7(3\times5)^{1/2}} & \dfrac{1}{3\times7(3\times5)^{1/2}} & \dfrac{1}{7(3\times5)^{1/2}}\\
0 & \dfrac{2}{3\times7\times5^{1/2}} & \dfrac{4}{7\times11\times5^{1/2}} & -\dfrac{2}{7\times11\times5^{1/2}}\\
0 & 0 & \dfrac{16}{3\times11(2\times3\times5\times7\times13)^{1/2}} & \dfrac{1}{11(2\times3\times5\times7\times13)^{1/2}}\\
0 & 0 & 0 & -\dfrac{1}{(2\times3\times7\times11\times13)^{1/2}}
\end{matrix}\right\}
\begin{pmatrix}C_2\\C_5\\C_8\\C_9\end{pmatrix}
=\frac{1}{(4\pi)^{1/2}}\begin{pmatrix}\Lambda_2\\\Lambda_5\\\Lambda_8\\\Lambda_9\end{pmatrix}. \qquad (69)
$$

---

[31] R. Vautier, thesis, University of Paris, 1954 (unpublished).

The matrices which effect the inverse transformations are, from Γ_α and Γ_γ.

$$\left\{\begin{matrix} 1 & 0 & -3\times3^{1/2} & 0 & \dfrac{(5\times7\times13)^{1/2}}{2\times2^{1/2}} \\[2ex] 0 & -\dfrac{5\times7^{1/2}}{2\times3^{1/2}} & 0 & \dfrac{-7(13)^{1/2}}{4\times2^{1/2}} & \dfrac{(7\times13)^{1/2}}{2(2\times5)^{1/2}} \\[2ex] 0 & 0 & \dfrac{7\times3^{1/2}}{2} & 0 & -\dfrac{3(5\times7\times13)^{1/2}}{2\times2^{1/2}} \\[2ex] 0 & 0 & 0 & \dfrac{7\times11(13)^{1/2}}{4\times2^{1/2}} & -\dfrac{11(7\times13)^{1/2}}{2(2\times5)^{1/2}} \\[2ex] 0 & 0 & 0 & 0 & \dfrac{11(7\times13)^{1/2}}{2(2\times5)^{1/2}} \end{matrix}\right\} \begin{bmatrix} \Lambda_1 \\ \Lambda_3 \\ \Lambda_4 \\ \Lambda_6 \\ \Lambda_7 \end{bmatrix} = (4\pi)^{1/2} \begin{bmatrix} C_1 \\ C_3 \\ C_4 \\ C_6 \\ C_7 \end{bmatrix}, \tag{70}$$

and from Γ_ϵ

$$\left\{\begin{matrix} (3\times5)^{1/2} & -(3/2)5^{1/2} & (2\times3\times5\times7\times13)^{1/2}/16 & (3/16)(2\times3\times7\times11\times13)^{1/2} \\ 0 & (3\times7\times5^{1/2})/2 & -(9/8)(2\times3\times5\times7\times13)^{1/2} & -(3/8)(2\times3\times7\times11\times13)^{1/2} \\ 0 & 0 & (3\times11/16)(2\times3\times5\times7\times13)^{1/2} & (3/16)(2\times3\times7\times11\times13)^{1/2} \\ 0 & 0 & 0 & -(2\times3\times7\times11\times13)^{1/2} \end{matrix}\right\} \begin{bmatrix} \Lambda_2 \\ \Lambda_5 \\ \Lambda_8 \\ \Lambda_9 \end{bmatrix} = (4\pi)^{1/2} \begin{bmatrix} C_2 \\ C_5 \\ C_8 \\ C_9 \end{bmatrix}. \tag{71}$$

7. NICKEL

Although the theory has been developed explicitly for an ionic model, one might hope that the general features would remain true for the case of a metal, and that the temperature dependence of the magnetoelastic contribution to the anisotropy would explain the extraordinarily rapid variation of the observed anisotropy of nickel. This is not the case. Becker and Döring[1] have estimated the contribution to the anisotropy from the external strains and found it to be an unimportant part of the total anisotropy of this material. Furthermore, because of their structure, neither nickel nor iron is capable of supporting internal strains which are even under inversion.

Although the magnetoelastic coupling seems incapable of accounting for the temperature dependence of the anisotropy of nickel, there is approximate agreement between the theoretical and the observed temperature dependence of the magnetostriction, as we now show.

The magnetostriction of a single crystal of nickel at room temperature was reported by Bozorth and Hamming[32] who fitted it to a five-constant series differing slightly from Eq. (67). Converting to the latter series, the results of Bozorth and Hamming are

$$\begin{gathered} C_1=-68.8\times10^{-6},\quad C_2=-73\times10^{-6}, \\ C_3=-7.8\times10^{-6},\quad C_4=-7.5\times10^{-6}, \\ C_5=15.4\times10^{-6}. \end{gathered} \tag{72}$$

On the basis of this result, Birss and Lee[33] measured the magnetostriction of nickel as a function of temperature and fitted their data to the series of Eq. (67), terminated at C_2. Their room temperature values are $C_1=-77.2\times10^{-6}$ and $C_2=-70.0\times10^{-6}$, in approximate agreement with Bozorth and Hamming. Lee and Birss[34] applied the analysis of Kittel and Van Vleck[23] to their measurements, and showed that the magnetostriction coefficients could be fitted by a polynomial in the magnetization with powers 3, 10 and 21, corresponding to $l=2$, 4, and 6 terms. While Lee and Birss plot their data as a function of temperature, we prefer to use the magnetization as the independent variable, and to employ the modified Bessel functions,[26] which are more appropriate at higher temperatures, while behaving properly at low temperatures.

Ignoring higher terms, we have from Eqs. (70) and (71) that

$$\begin{gathered} \Lambda_1 \cong (4\pi)^{1/2}C_1, \\ (3\times5)^{1/2}\Lambda_2 \cong (4\pi)^{1/2}C_2. \end{gathered} \tag{73}$$

Furthermore, from Eqs. (66a) and (66b),

$$\begin{gathered} \Lambda_1=(1/c_0{}^\gamma)\bar{B}_{0,2}{}^\gamma, \\ \Lambda_2=(1/c_0{}^\epsilon)\bar{B}_{0,2}{}^\epsilon. \end{gathered} \tag{74}$$

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[32] R. M. Bozorth and R. W. Hamming, Phys. Rev. **89**, 865 (1953).

[33] R. R. Birss and E. W. Lee, Proc. Phys. Soc. (London) **76**, 502 (1960).

[34] E. W. Lee and R. R. Birss, Proc. Phys. Soc. (London) **78**, 391 (1961).

Then, from Eqs. (15) and (52),

$$\Lambda_1 = \frac{5}{2(c_{11}-c_{12})} B_{0,2}{}^{\gamma} \hat{I}_{5/2}\left(3\frac{Tc}{T}m_0\right), \tag{75a}$$

$$\Lambda_2 = \frac{1}{c_{44}} B_{0,2}{}^{\epsilon} \hat{I}_{5/2}\left(3\frac{Tc}{T}m_0\right). \tag{75b}$$

Thus

$$\frac{(c_{11}-c_{12})(T)}{(c_{11}-c_{12})(0)} \frac{C_1(T)}{C_1(0)} \cong \hat{I}_{5/2}\left(3\frac{Tc}{T}m_0\right), \tag{76a}$$

and

$$\frac{c_{44}(T)}{c_{44}(0)} \frac{C_2(T)}{C_2(0)} \cong \hat{I}_{5/2}\left(3\frac{Tc}{T}m_0\right), \tag{76b}$$

as, at $T=0°\text{K}$,

$$\hat{I}_{(2l+1)/2}(\infty) = 1. \tag{77}$$

The elastic constants of nickel have been measured by Alers, Neighbors, and Sato.[35] To express Eqs. (76a) and (76b) in terms of the magnetization we employ the magnetothermal measurements of Pugh and Argyle,[36] Foner and Thompson,[37] and P. Weiss,[38] in complementary temperature ranges.

In Fig. 2 we plot

$$[(c_{11}-c_{12})(T)/(c_{11}-c_{12})(0)][C_1(T)/C_1(0)]$$

as a function of the experimental magnetization in curve (a) and

$$[c_{44}(T)/c_{44}(0)][C_2(T)/C_2(0)]$$

in curve (b), combining the measurements of the elastic constants, magnetostriction and magnetization, and eliminating the temperature explicitly. On the same figure, as curve (c), we also show the modified Bessel function $I_{5/2}/I_{1/2}$ as a function of the Langevin magnetization. Birss and Lee found a broad maximum in $|C_1|$ versus T which is only partially reduced by the temperature dependence of the elastic constant $(c_{11}-c_{12})$. The Γ_{ϵ} magnetostriction coefficient also shows evidence of the mixing in of a higher degree term at low temperatures. While the theoretical curve has an initial slope of 3, curve (b) initially drops approximately in accordance with the tenth-power law. At room temperature nickel has a reduced magnetization of about 0.935, and from Fig. 1, at this magnetization $\hat{I}_{5/2}\cong 0.8$ and $\hat{I}_{9/2}\cong 0.5$. Thus, from the data of Bozorth and Hamming and on the basis of the preceding analysis one might expect that at 0°K,

$$\begin{aligned} C_1(0)&\cong -73\times 10^{-6}, & C_2(0)&\cong -87\times 10^{-6}, \\ C_4(0)&\cong -14\times 10^{-6}, & C_5(0)&\cong 29\times 10^{-6}, \end{aligned} \tag{78}$$

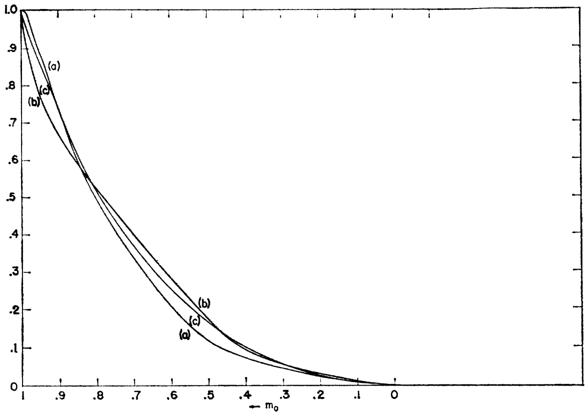

FIG. 2. (a) Experimental magnetostriction times elastic constant (Γ_{γ}) vs experimental magnetization. See Eq. (76a) of text. (b) experimental magnetostriction times elastic constant (Γ_{ϵ}) vs experimental magnetization. See Eq. (76b) of text. (c) theoretical $\hat{I}_{5/2}$ vs m_0. Elastic Constants: Alers, Neighbours, and Sato, reference 35; Magnetostriction: Birss and Lee, reference 33; Magnetization: Pugh and Argyle, reference 36; Foner and Thompson, reference 37; P. Weiss, reference 38.

with the corresponding magnetoelastic coupling coefficients

$$\begin{aligned} B_{0,2}{}^{\gamma} &\cong -(4\pi)^{1/2} 38\times 10^6 \text{ ergs/cm}^3, \\ B_{0,2}{}^{\epsilon} &\cong -(4\pi)^{1/2} 28\times 10^6 \text{ ergs/cm}^3, \\ B_{0,4}{}^{\gamma} &\cong -(4\pi)^{1/2} 1.0\times 10^6 \text{ ergs/cm}^3, \\ B_{0,4}{}^{\epsilon} &\cong (4\pi)^{1/2} 1.6\times 10^6 \text{ ergs/cm}^3. \end{aligned} \tag{79}$$

Magnetostriction measurements in progress at the Naval Ordnance Laboratory will determine if the inclusion of the higher degree terms does indeed resolve the deviations from the theory, particularly in the case of the Γ_{ϵ} terms.

Employing these coefficients in Eq. (21b), one finds the magnetoelastic contribution to be a negligible fraction of the fourth degree anisotropy of nickel at 0°K, and to be of the correct sign but still too small to account for the change in sign of κ_4 of nickel at high temperatures.

8. THE SYMMETRY OF THE DISTORTED CRYSTAL

Of the 48 symmetry operators of the cubic group many are destroyed by the external or internal distortions produced by the magnetoelastic coupling. These distortions generally constitute small perturbations on an essentially cubic structure. The symmetry of these distortions, in principle detectable by x rays, provides information on the magnetoelastic coupling in the crystal.

The inversion operation is an element of the original cubic group, and it remains an element of the distorted crystal. We, therefore, need only consider the 24 proper rotations of the cubic group. These 24 symmetry elements are listed in Table V. We also list the ten types of strain components; ϵ^{α}; $\epsilon^{\beta'}$; $\epsilon_1{}^{\gamma}$; $\epsilon_2{}^{\gamma}$; $\epsilon_1{}^{\epsilon}$; $\epsilon_2{}^{\epsilon}$; $\epsilon_3{}^{\epsilon}$;

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TABLE V. Symmetry elements of the cubic group. A plus sign indicates that the basis function goes into plus itself under the particular operation.

		Γ_α	$\Gamma_{\beta'}$	Γ_γ (1)	Γ_γ (2)	Γ_ϵ x	Γ_ϵ y	Γ_ϵ z	$\Gamma_{\delta'}$ x	$\Gamma_{\delta'}$ y	$\Gamma_{\delta'}$ z
E		+	+	+	+	+	+	+	+	+	+
C_2: x through	(π)	+	+	+	+	+			+		
C_2: y	(π)	+	+	+	+		+			+	
C_2: z	(π)	+	+	+	+			+			+
C_4: x	($\pi/2$)	+							+		
C_4: x	($-\pi/2$)	+							+		
C_4: y	($\pi/2$)	+								+	
C_4: y	($-\pi/2$)	+								+	
C_4: z	($\pi/2$)	+		+							+
C_4: z	($-\pi/2$)	+		+							+
C_2: [011]	(π)	+				+					
C_2: $[01\bar{1}]$	(π)	+				+					
C_2: [101]	(π)	+					+				
C_2: $[10\bar{1}]$	(π)	+					+				
C_2: [110]	(π)	+		+				+			
C_2: $[1\bar{1}0]$	(π)	+		+				+			
C_3: [111]	($2\pi/3$)	+	+								
C_3: [111]	($-2\pi/3$)	+	+								
C_3: $[1\bar{1}1]$	($2\pi/3$)	+	+								
C_3: $[1\bar{1}1]$	($-2\pi/3$)	+	+								
C_3: $[1\bar{1}\bar{1}]$	($2\pi/3$)	+	+								
C_3: $[1\bar{1}\bar{1}]$	($-2\pi/3$)	+	+								
C_3: $[11\bar{1}]$	($2\pi/3$)	+	+								
C_3: $[11\bar{1}]$	($-2\pi/3$)	+	+								
J inversion		+	+	+	+	+	+	+	+	+	+

$\epsilon_1^{\delta'}$; $\epsilon_2^{\delta'}$; $\epsilon_3^{\delta'}$. For each such strain we indicate by a + those symmetry operations which leave it invariant.

If the crystal supports an internal strain of type $\epsilon^{\beta'}$ then the distorted crystal will be invariant only under those operations which leave $\epsilon^{\beta'}$ invariant; in this case E, $3C_2$, $8C_3$, J, $3JC_2$, $8JC_3$. Thus, the crystal will have the point group T_h [or $(2/m)\bar{3}$]—provided, of course, that no other distortions are present.

If several types of distortion are simultaneously present the only symmetry elements which survive are those to which every distortion is symmetric; that is, only those operations in Table V which have a + under every nonzero distortion. Thus, if both $\Gamma_{\beta'}$ and Γ_γ distortions are induced the surviving symmetry operations are E, $3C_2$, J, $3JC_2$. The crystal then has the point group D_{2h} [or $(2/m)(2/m)(2/m)$].

In the above discussion, it is presumed that the several ϵ's in a multidimensional representation are unequal. However, if, for example, $\epsilon_1^\epsilon=\epsilon_2^\epsilon=\epsilon_3^\epsilon\neq 0$ additional symmetries may appear, which cannot be analyzed simply on the basis of Table V. In this case the procedure for finding the surviving symmetries would be to write the full 3×3 matrix for each symmetry operation in the Γ_ϵ representation, and to find which of these have the vector (1,1,1) as an eigenvector with an eigenvalue of +1. Doing this explicitly we find that the surviving operations are the twofold rotations around the $[01\bar{1}]$, $[10\bar{1}]$, and $[1\bar{1}0]$ axes, the threefold rotations around [111], the identity, and the product of all of these operations with the inversion. The corresponding point group is D_{3d} [or $\bar{3}(2/m)$].

Of particular interest are those specific directions of the (sublattice) magnetization for which certain distortions vanish. The crystal is then more symmetric than it is for arbitrary directions of the (sublattice) magnetization.

Consider the one-dimensional representation $\Gamma_{\beta'}$. The distortion $\epsilon^{\beta'}$ is proportional to a sum of Kubic harmonics $K^{\beta' l}$, for various l. But $K^{\beta' l}$ has six nodal planes; all those planes containing a face diagonal of the cube and the axis perpendicular to that face diagonal. Hence, the distortion $\epsilon^{\beta'}$ vanishes if the (sublattice) magnetization lies in any of these planes.

Similarly ϵ_2^γ vanishes if the (sublattice) magnetization lies in either of the two planes containing the z axis and one of the face-diagonals perpendicular to it.

The strain ϵ_3^ϵ vanishes if the (sublattice) magnetization lies in either the x-z plane or the y-z plane.

And finally $\epsilon_3^{\delta'}$ vanishes if the (sublattice) magnetization lies in any of the four planes containing the z axis and either a face-diagonal or a cube axis perpendicular to the z axis.

The locus of the directions of (sublattice) magnetization for which other strain components (such as $\epsilon_1^{\delta'}$) vanish can be obtained from the above by a simple permutation of the coordinates.

We now examine the particular crystal symmetry which results if the (sublattice) magnetization lies along one of the principal symmetry directions of the cubic system; that is, along [111], [001], or [110].

We first consider the (sublattice) magnetization to lie along the cube diagonal [111]. This direction lies in the nodal planes for $\epsilon^{\beta'}$, ϵ_1^γ, ϵ_2^γ, $\epsilon_1^{\delta'}$, $\epsilon_2^{\delta'}$, and $\epsilon_3^{\delta'}$. Furthermore, examination of the Kubic harmonics of Γ_ϵ shows that $\epsilon_1^\epsilon=\epsilon_2^\epsilon=\epsilon_3^\epsilon$. This is a symmetry which we have discussed previously, the resulting point group being D_{3d} [or $\bar{3}(2/m)$].

If the (sublattice) magnetization lies in the [001] direction it lies in the nodal planes of $\epsilon^{\beta'}$, ϵ_2^γ, ϵ_1^ϵ, ϵ_2^ϵ, ϵ_3^ϵ, $\epsilon_1^{\delta'}$, $\epsilon_2^{\delta'}$, and $\epsilon_3^{\delta'}$. That is, only ϵ_1^γ can be nonzero. The surviving symmetry elements are, from Table V, the identity, the three twofold rotations around the cube axes, the fourfold rotation around the z axis, the twofold rotations around [110] and $[1\bar{1}0]$, and the product of each of these with the inversion. The point group is D_{4h} [or $(4/m)(2/m)(2/m)$].

Finally, consider the (sublattice) magnetization to be in the [110] direction. This direction lies in the nodal planes of $\epsilon^{\beta'}$, ϵ_2^γ, ϵ_1^ϵ, ϵ_2^ϵ, $\epsilon_1^{\delta'}$, and $\epsilon_2^{\delta'}$. Thus, only ϵ_1^γ, ϵ_3^ϵ, and $\epsilon_3^{\delta'}$ can be nonzero. From Table V we see that these three strain components are simultaneously symmetric only to E and to the twofold rotation around the z axis (and the product of these with the inversion). Hence, the point group has only the very low symmetry of C_{2h} (or $2/m$). If the particular crystal did not support an internal strain belonging to $\Gamma_{5'}$, however, the additional symmetry elements of twofold rotation around [110] and [1$\bar{1}$0] would survive, and the point group would be D_{2h} [or $(2/m)(2/m)(2/m)$]. Thus, examination of the crystal symmetry, by x-ray diffraction, as a function of the direction of (sublattice) magnetization can give interesting information as to the magnetoelastic coupling.

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Infrared Light Stimulation and Quenching in ZnS Phosphors*

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The effects of infrared on the luminescence of three ZnS phosphors (activated with Cu, Ag, and Cu-Pb) at liquid nitrogen temperatures has been investigated. The transient stimulation and permanent quenching (or enhancement) was determined at various wavelengths in the emission spectrum of the ultraviolet-excited phosphors. Two infrared bands were used, one at about 0.75, the other at about 1.3. The effect of the infrared varies with the emission wavelength but not sufficiently to explain discrepancies with the usually accepted phosphor model. A modification of this model consisting of a coupled trapped electron-ionized activator complex is proposed, and the consequences are discussed.

I. INTRODUCTION

In a previous paper (hereafter called I), Kallmann and Luchner[1] have reported on measurements concerning the mechanism of ir (infrared) light stimulation in ZnS-type phosphors. The main result was that such stimulation cannot be brought about by an independent direct release of trapped electrons as was often assumed. It was shown that many effects concerning stimulation could be understood with the assumption that the ir somehow produces a faster recombination between conduction electrons and ionized activators, which leads to a transient increase in luminescent intensity. In order to provide a model for stimulation and quenching, it is necessary to find out more about the mechanism by which the ir produces faster recombinations and quenching at the same time, as indicated by many experiments. The present paper deals with this question and proposes a model for trapping of electrons and this release by ir which is somewhat different than envisaged up to now.

Before the experiments and their interpretation are given, we will summarize numerous discrepancies between the "old" model which has been rather successfully used up to now,[2] and results already obtained.

(1) Both stimulation and quenching of luminescence and photoconductivity by infrared evidence the same infrared wavelength dependence[3–6] showing that they are produced by the same elementary process. This cannot be understood by using the assumption that stimulation is due to the independent release of electrons from traps and that quenching is due to the independent release of holes from ionized activators.

(2) The equilibrium quenching of luminescence (due to infrared) is less than that of the photoconductivity[7]; if the light emission is proportional to the product of n and P_t (see paragraph 4 below), light quenching should be greater.

(3) Infrared stimulation of luminescence after excitation is not instantaneous but has a finite rise[8]; this

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