

Theory of the Temperature Dependence of the Magnetoelastic Constants of Cubic Crystals

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The conventional theory of the variation with temperature of the anisotropy of cubic crystals is extended to include the magnetoelastic constants.

AT the Grenoble conference, one of us¹ reported a derivation of the relation $K_1 \sim M^{10}$ between the anisotropy constant K_1 and saturation magnetization M of cubic crystals which was somewhat more general than previous proofs. The other of us² commented that the same method could be used to derive a relation between the magnetoelastic constants and the magnetization or temperature. It is the purpose of the present note to do this and also to discuss the conditions under which the result is valid. As is customary in the discussion of anisotropy and related subjects, our results will be expressed in the form of a functional relation connecting the magnetoelastic energy with M rather than T , but as the saturation magnetization M is itself a function of T , the form of dependence on T is in principle determined.

Becker and Döring³ show by symmetry considerations that in a cubic crystal, the magnetoelastic energy is given by an expression of the form

$$F_A^0 = \beta_0[A_{11} + A_{22} + A_{33}] + \beta_1[(\alpha_1^2 - \frac{1}{3})A_{11} + (\alpha_2^2 - \frac{1}{3})A_{22} + (\alpha_3^2 - \frac{1}{3})A_{33}] + \beta_2[2\alpha_1\alpha_2A_{12} + 2\alpha_2\alpha_3A_{23} + 2\alpha_3\alpha_1A_{13}] + \beta_3s'(A_{11} + A_{22} + A_{33}) + \beta_4[p_4(\alpha_1)A_{11} + p_4(\alpha_2)A_{22} + p_4(\alpha_3)A_{33}] + \beta_5[(7\alpha_1\alpha_2\alpha_3^2 - \alpha_1\alpha_2)A_{12} + (7\alpha_2\alpha_3\alpha_1^2 - \alpha_2\alpha_3)A_{23} + (7\alpha_3\alpha_1\alpha_2^2 - \alpha_3\alpha_1)A_{13}], \quad (1)$$

with

$$p_4(\mu) = (8/35)P_4(\mu) = [\mu^4 - (6/7)\mu^2 + (6/70)], \\ s' = \alpha_1^2\alpha_2^2 + \alpha_1^2\alpha_3^2 + \alpha_2^2\alpha_3^2 - \frac{1}{5}.$$

The direction cosines of the magnetization relative to the principal cubic axes are denoted by $\alpha_1, \alpha_2, \alpha_3$, and the strain tensor A_{ij} is defined by

$$X = x_0 + A_{11}x + A_{12}y + A_{13}z, \text{ etc. } (A_{ij} = A_{ji}).$$

¹ J. H. Van Vleck, *J. phys. radium* **20**, 128 (1959). See this article for earlier references and a review of the theory of ferromagnetic anisotropy, with which it is assumed the reader has some familiarity.

² C. Kittel, *J. phys. radium* **20**, 135 (1959).

³ R. Becker and W. Döring, *Ferromagnetismus* (Verlag-Julius Springer, Berlin, 1939), p. 136.

The constants β_0, \dots, β_5 which we use are connected with the b constants used by Becker and Döring as follows

$$\beta_0 = b_0 + \frac{1}{5}b_3; \quad \beta_1 = b_1 + (6/7)b_4; \quad \beta_2 = b_2 + (1/7)b_5, \\ \beta_3 = b_3 + \frac{2}{5}b_4; \quad \beta_4 = b_4; \quad \beta_5 = (2/7)b_5.$$

We believe our grouping of terms or definitions of constants is more fundamental as well as convenient, since thereby terms with different modes of temperature variation are sorted out. In particular, the coefficient of each β is a homogeneous surface harmonic, of degree 4 for $\beta_3, \beta_4, \beta_5$, and of the second degree for β_1, β_2 . The coefficients of the Becker and Döring b_3, b_4, b_5 , on the other hand, are mixtures of harmonics of degree 0, 2, 4, and we have absorbed the portions of the 0th and 2nd degree in $\beta_0, \beta_1, \beta_2$.

The factors β_0, \dots, β_5 or b_0, \dots, b_5 , can be functions of the magnetization or equivalently of temperature. In order to have more useful constants, $\beta'_0, \dots, \beta'_5$, which do not involve these variables, we have simply to repeat Becker and Döring's symmetry argument under the assumption that the local magnetization of a constituent has a modulus independent of temperature so that thermal effects arise only from its degree of alignment. By a constituent we mean merely a single atom in the one-atom crystalline potential model of anisotropy, or each appreciably coupled atom-pair in the model in which anisotropy arises from modulations of interatomic coupling. We use the term local magnetization for the resultant moment of an atom or atom-pair. If the magnetism arises entirely from spin, the assumption of a constant local magnetization requires that $(S_{x1})^2 + (S_{y1})^2 + (S_{z1})^2$ in the one-atom, or $(S_{x1} + S_{x2})^2 + (S_{y1} + S_{y2})^2 + (S_{z1} + S_{z2})^2$ in the atom-pair, be treated as an absolute constant. The Becker and Döring symmetry argument shows that now the expression for the magnetoelastic energy can also be written as

$$F_A^0 = \beta'_0[A_{11} + A_{22} + A_{33}] + \beta'_1[\langle \alpha_1^{(i)^2} - 1 \rangle_{av} A_{11} + \dots] + \dots + (2/7)\beta'_5 \times [\dots + \langle 7\alpha_3^{(i)}\alpha_1^{(i)}\alpha_2^{(i)^2} - \alpha_3^{(i)}\alpha_1^{(i)} \rangle_{av} A_{13}], \quad (2)$$

instead of (1). Here the average is to be taken over all the constituents, which can differ in the direction of

their local magnetization specified by the direction cosines.

We now use the transformation theorem for normalized Tesselar harmonics,⁴

$$T_1^{m'}(\theta', \varphi') = \sum_m D_{m'm}(\Theta, \Phi, \Psi) T_1^m(\theta, \varphi), \quad (3)$$

taking the unprimed polar axis along the direction of the resultant magnetization of the crystal, and the axes for the primed system fixed relative to the principal cubic axes. The Eulerian angles connecting the two systems are Θ, Φ, Ψ . If we regard all anisotropic effects as perturbations, so that they do not need to be included in the computation of the average values, then there is axial symmetry about the direction of magnetization in the unperturbed system, and only the member $m=0$ of (3) averages to anything different from zero. Furthermore, expect for a normalization factor, $D_{m'0}$ is an ordinary Tesselar harmonic. Hence

$$\langle T_1^{m'}(\theta', \varphi') \rangle_{av} = T_1^{m'}(\Theta, \Phi) \langle P_1(\cos\theta) \rangle_{av},$$

where the Legendre polynomials $P_l(\mu)$ are normalized in the usual way, so that they reduce to unity at the poles.

From (3) we see that

$$\langle \alpha_1^{(i)2} - \frac{1}{3} \rangle_{av} = (\alpha_1^2 - \frac{1}{3}) \langle P_2(\cos\theta) \rangle_{av} \text{ etc.} \quad (4)$$

The angle θ is that between the local magnetization and the macroscopic magnetization M .

If zero subscripts denote the values at $T=0$, then if one is close to saturation,

$$\langle P_1(\cos\theta) \rangle_{av} = (M/M_0)^{l(l+1)/2} \quad (5)$$

as has been shown by various writers, with progressively increasing generality, beginning with Akulov.^{5,1}

On comparing (1), and (2), using (4), (5) and remembering that the β_i' are independent of M , we obtain our final result

$$\begin{aligned} \frac{\beta_0}{\beta_{00}} = 1, \quad \frac{\beta_1}{\beta_{10}} = \frac{\beta_2}{\beta_{20}} = \left(\frac{M}{M_0} \right)^3, \\ \frac{\beta_3}{\beta_{30}} = \frac{\beta_4}{\beta_{40}} = \frac{\beta_5}{\beta_{50}} = \left(\frac{M}{M_0} \right)^{10}. \end{aligned} \quad (6)$$

It remains only to comment on the conditions under which (6) may be expected really to apply. Thermal expansion has been neglected in treating the β_i' as absolute constants. The one-atom model can contribute to $\beta_3, \beta_4, \beta_5$ only if the spin of atom-atom exceeds $\frac{3}{2}$. In the two-atom model it must be supposed that the resultant spin of the pair has its maximum value, in other words there must be what Keffer⁵ calls perfect correlation. With incomplete correlation the dependence on temperature or magnetization will be less drastic. In the atom-pair model, the fourth order effects can arise either from the first order effect of anisotropic exchange of quadrupolar form or the second order effect of that of dipolar form. Keffer⁶ has corrected the work of various writers, and shown that the latter gives the same form of temperature dependence as the former. Regardless of the model used, it is basic to the calculation in obtaining (6) that the $(M_0 - M)/M_0$ be small compared with unity. The appropriate curve replacing (5) at higher temperatures has been given by Yosida and Tachiki⁷ for the case $l=4$ for the one-atom or completely correlated atom-pair model.

Experimentally, the place par excellence to test the theory is, of course, the ferrites or yttrium iron garnet, as questions of electron migration do not arise, and a considerable part of the anisotropy arises from the one-atom model. No adequate experimental data are available, but the theory is very similar to that for the temperature variation of the anisotropy, and the latter is well confirmed for the Mn^{++} and Fe^{+++} ions in the ferrites. On the other hand, in metallic iron or nickel, the experimental discrepancies relative to the tenth power relation predicted by theory are notoriously bad.⁸ As the way in which the directional factors enter is rather similar in the fourth order part of the magnetostriction and of the anisotropy constant K_1 , it is perhaps a fair approximation to assume that $\beta_3, \beta_4, \beta_5$ depend on temperature in approximately the same way as does K_1 , and that β_1, β_2 vary considerably less rapidly.

⁴ E. P. Wigner, *Group Theory and Its Application to Atomic Spectra* (Academic Press, Inc., New York and London, 1959), Chap. 15.

⁵ N. Akulov, Z. Physik **100**, 197 (1936); C. Zener, Phys. Rev. **96**, 1335 (1954); F. Keffer, Phys. Rev. **100**, 1692 (1955); J. H. Van Vleck, see reference one.

⁶ F. Keffer and T. Oguchi, Phys. Rev. **117**, 718 (1960).

⁷ K. Yosida and M. Tachiki, Progr. Theoret. Phys. **17**, 331 (1957).

⁸ C. D. Graham, Jr., *Fifth Conference on Magnetism and Magnetic Materials, Detroit, Michigan, November, 1959* [J. Appl. Phys. (to be published)], paper 55.