

Classical Theory of the Temperature Dependence of Magnetic Anisotropy Energy*

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The consequences are analyzed of the following two assumptions: (1) the effect of temperature upon magnetic anisotropy arises solely from the introduction of local deviations in the direction of magnetization; and (2) the local deviation in an elementary region is the resultant of a very large number of independent deviations. The influence of these local deviations upon the magnetic anisotropy is most conveniently expressed by representing the magnetic energy as a series of surface harmonics. The coefficient of the n th harmonic is found to vary with temperature as $\{J_s(T)/J_s(0)\}$ raised to the power $n(n+1)/2$. The first two exponents for cubic crystals have values of 10 and 21, respectively. The exponent 10 expresses almost precisely the observed temperature dependence of K_1 in iron. In nickel the anisotropy decreases much more rapidly than predicted. It is deduced that the above two assumptions are applicable to iron but not to nickel.

1. INTRODUCTION AND RESULTS

THE magnetization energy of a ferromagnetic crystal is usually dependent to some extent upon the relative orientation of the magnetization vector with respect to the crystalline axes. It is customary to describe this anisotropy of magnetic energy in terms of the coefficients in a Fourier-like expansion of the magnetic energy. Thus, K_1, K_2, \dots , are the first, second, \dots , coefficients which symmetry requirements do not require to be precisely zero. As an example,

$$E_{\text{mag}} = E_0 + K_1(\alpha_1^2\alpha_2^2 + \alpha_2^2\alpha_3^2 + \alpha_3^2\alpha_1^2) + K_2\alpha_1^2\alpha_2^2\alpha_3^2 + \dots \quad (1)$$

for a crystal with cubic symmetry, and

$$E_{\text{mag}} = E_0 + K_1 \sin^2\theta + K_2 \sin^4\theta + \dots \quad (2)$$

for a crystal with hexagonal symmetry. Here $\alpha_1, \alpha_2, \alpha_3$ are the cosines of the magnetization vector with respect to the cubic axes, and θ is the angle which the magnetization vector makes with the hexagonal axis.

As is illustrated in Fig. 1 for iron, cobalt, and nickel, the anisotropy coefficients of metals decrease very rapidly with increasing temperature. One is tempted to interpret this marked temperature dependence somewhat as follows: the anisotropy energy must have its origin in a coupling between the spins and the orbital motion, which in turn is coupled to the crystallographic axes, and we then interpret the temperature dependence of the anisotropy coefficients as simply a manifestation of the gradual weakening of this coupling with rising temperature. The fallacy of this naive interpretation is evident upon observing the temperature dependence of the saturation magnetostriction, λ_s . Magnetostriction also arises from a spin-orbit coupling. It decreases with temperature but at a much slower rate than does anisotropy. The fallacy of this naive interpretation is also evident upon observing that the deviation of g from 2 is a measure of the spin-orbit coupling, and

experimentally this deviation is found to be essentially constant in nickel up to the Curie temperature.¹

This paper presents an attempt to understand this marked temperature dependence of the crystal anisotropy. In this attempt we proceed in a classical manner, no quantum mechanical effects being introduced. Two basic assumptions are made. The first assumption is that the sole effect of temperature is to introduce local fluctuations in the direction of the magnetization vector \mathbf{J} . This assumption implies that the local magnitude of \mathbf{J} is independent of temperature, and that Eqs. (1) and (2) represent correctly the local density of magnetization energy, the coefficients K_1, K_2, \dots being independent of temperature. The second assumption is that the deviation in the direction of the local \mathbf{J} from the direction of the macroscopic \mathbf{J} is the resultant of a large number of very small deviations having independently random directions. This second assumption is in accord with the spin wave description of the influence of temperature upon magnetization, each independent small deviation being associated with a particular spin wave. A consequence of this second assumption is that the probability that the angle θ between the local and the macroscopic \mathbf{J} lie within a certain range is given by a random walk function appropriate to a spherical surface.

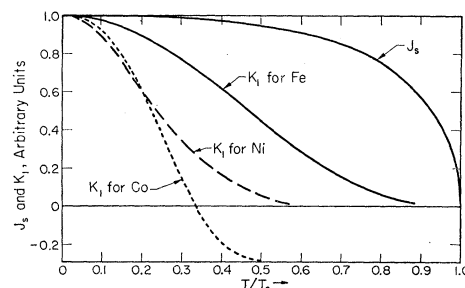


Fig. 1. Temperature dependence of anisotropy energy in Fe, Co, and Ni.

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¹ N. Bloembergen, Phys. Rev. **78**, 572 (1950); Bagguley and Harrick, Proc. Phys. Soc. (London) **A67**, 648 (1954).

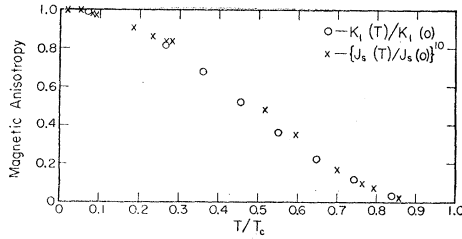


FIG. 2. Comparison of experimental and theoretical temperature dependence of K_1 for iron. Experimental values of $K_1(T)$ from Bozorth (see reference 2). Experimental data for $J_s(T)/J_s(0)$ at low temperature after Bozorth (see reference 3), at high temperatures after Potter (see reference 4).

The consequences of the above assumptions are analyzed in Sec. (2). In order to be able to express the results succinctly, we rewrite Eqs. (1) and (2) in the form

$$E_{\text{mag}} = \sum_n E_n S_n(\alpha_1, \alpha_2, \alpha_3), \quad (3)$$

where S_n is a surface harmonic of order n having the appropriate symmetry. The result of this analysis is that $E_{\text{mag}}(\alpha_1, \alpha_2, \alpha_3)$ averaged over the random walk function satisfies an equation identical to Eq. (3) except that now $\alpha_1, \alpha_2, \alpha_3$ refer to the direction cosines of the macroscopic \mathbf{J} , and the coefficients $E_n(T)$ are related to the original coefficients $E_n(0)$ by the relation

$$E_n(T)/E_n(0) = \{J_s(T)/J_s(0)\}^{n(n+1)/2}, \quad (4)$$

where J_s is the magnetic saturation. In particular,

$$E_4(T)/E_4(0) = \{J_s(T)/J_s(0)\}^{10}. \quad (5)$$

Since

$$S_4(\alpha_1, \alpha_2, \alpha_3) = \alpha_1^2 \alpha_2^2 + \alpha_2^2 \alpha_3^2 + \alpha_3^2 \alpha_1^2 - \frac{1}{3},$$

we are tempted to compare theory with experiment²⁻⁴ by comparing the right member of Eq. (5) with the observed value of $K_1(T)/K_1(0)$ for a crystal with cubic symmetry. Such a comparison is made for iron in Fig. 2. The essentially perfect agreement indicates that our two basic assumptions correspond rather closely to reality, at least for this metal.

Upon reference to Fig. 1, we see that K_1 decreases at least twice as rapidly with T/T_c in the case of nickel as in the case of iron. Since J_s decreases in approximately the same manner for these two metals, we conclude that the same agreement will not be obtained for nickel as was obtained for iron. In seeking for an interpretation of this disagreement, we observe that the expression $\alpha_1^2 \alpha_2^2 + \alpha_2^2 \alpha_3^2 + \alpha_3^2 \alpha_1^2$ occurs not only in $S_4(\alpha_1, \alpha_2, \alpha_3)$ but also in $S_6(\alpha_1, \alpha_2, \alpha_3)$. Thus, we find that

$$S_6(\alpha_1, \alpha_2, \alpha_3) = \alpha_1^2 \alpha_2^2 \alpha_3^2 - (1/11)(\alpha_1^2 \alpha_2^2 + \alpha_2^2 \alpha_3^2 + \alpha_3^2 \alpha_1^2) + (2/231).$$

If E_6 were the first nonvanishing coefficient, other than E_0 , in the expansion of Eq. (3), the coefficient of

$\alpha_1^2 \alpha_2^2 + \alpha_2^2 \alpha_3^2 + \alpha_3^2 \alpha_1^2$ would vary as the 21st power of $J_s(T)/J_s(0)$. Such a variation would indeed agree quite well with the observed variation of $K_1(T)/K_1(0)$. Such an interpretation would, of course, imply a large value of the coefficient $K_2(0)$ compared to $K_1(0)$. The writer is indebted to Dr. H. Sato of this laboratory for a careful analysis of the low-temperature data for nickel,⁵ from which analysis one must conclude that $K_2(0)$ cannot be sufficiently large compared to $K_1(0)$ to render acceptable the above interpretation of the discrepancy for this metal. We must conclude that our two basic assumptions, while valid for iron, are invalid for nickel.

Two attempts have been made to compute the temperature dependence of K_1 by quantum mechanical methods. The first, by Akulov,⁶ proceeds somewhat in the spirit of the present paper. He found that at low temperatures, $K_1(T)/K_1(0)$ decreased ten times as rapidly as $J_s(T)/J_s(0)$, and recognized that a reasonable generalization of this variation was a tenth power law. He showed that the experimental data for iron were consistent with a power of 10.03. The second attempt was by Van Vleck.⁷ His computed values of $K_1(T)/K_1(0)$ varied between the fifth and sixth power of $J_s(T)/J_s(0)$.

2. ANALYSIS

In this section, our objective is to compute the average of $S_n(\alpha_1, \alpha_2, \alpha_3)$ with respect to the appropriate random walk function. The origin of the random walk will be taken at $(\bar{\alpha}_1, \bar{\alpha}_2, \bar{\alpha}_3)a$ on the surface of a sphere of radius a .

The random walk distribution function will have symmetry about $(\bar{\alpha}_1, \bar{\alpha}_2, \bar{\alpha}_3)$. We shall, therefore, find the average of $S_n(\alpha_1, \alpha_2, \alpha_3)$ as the direction $(\alpha_1, \alpha_2, \alpha_3)$ describes a cone about $(\bar{\alpha}_1, \bar{\alpha}_2, \bar{\alpha}_3)$. Towards this end, we let θ be the polar coordinates of $(\alpha_1, \alpha_2, \alpha_3)$ with respect to $(\bar{\alpha}_1, \bar{\alpha}_2, \bar{\alpha}_3)$. We then expand

$$S_n(\alpha_1, \alpha_2, \alpha_3) = \sum_{m=-n}^n C_m Y_n^m(\theta, \varphi). \quad (6)$$

When we now take the average of both sides with respect to φ , all terms vanish on the right side except that corresponding to $m=0$.

$$\langle S_n(\alpha_1, \alpha_2, \alpha_3) \rangle_\varphi = C_0 P_n(\cos\theta). \quad (7)$$

The constant C_0 is found by giving θ the particular value of zero. We thereby obtain

$$\langle S_n(\alpha_1, \alpha_2, \alpha_3) \rangle_\varphi = P_n(\cos\theta) S_n(\bar{\alpha}_1, \bar{\alpha}_2, \bar{\alpha}_3). \quad (8)$$

Our second and final step is to obtain the average of $P_n(\cos\theta)$ with respect to the appropriate random walk distribution function. This distribution is a function of

² R. M. Bozorth, *Ferromagnetism* (D. Van Nostrand Company, Inc., New York, 1951), pp. 569-578.

³ Reference 2, p. 720.

⁴ H. H. Potter, Proc. Roy. Soc. (London) **146**, 362 (1934).

⁵ Honda, Masumoto, and Shirakawa, Sci. Rpts. Tohoku Imp. Univ. **24**, 391 (1935-36).

⁶ N. Akulov, Z. Physik **100**, 197 (1936).

⁷ J. H. Van Vleck, Phys. Rev. **52**, 1178 (1937).

θ and of some parameter τ which determines the spread of the distribution function. Upon recalling that diffusion is a random walk process, we recognize that the distribution function $RW(\theta, \tau)$ will obey the standard diffusion differential equation if we associate τ with time, or more appropriately, with the dimensionless quantity tD/a^2 , where D is the diffusion coefficient, a the radius of the spherical surface upon which diffusion is imagined to be occurring. Thus, we have

$$\frac{\partial}{\partial \tau} RW(\theta, \tau) = \frac{1}{\sin \theta} \frac{\partial}{\partial \theta} \sin \theta \frac{\partial}{\partial \theta} RW(\theta, \tau). \quad (9)$$

The solution of this differential equation, subject to the boundary condition that $RW(\theta, \tau)$ approaches a delta function about $\theta=0$ as $\tau \rightarrow 0$, is

$$RW(\theta, \tau) = \sum_{n=0}^{\infty} \left\{ \int_{-1}^1 P_n^2(\cos \theta) d \cos \theta \right\}^{-1} \times e^{-n(n+1)\tau} P_n(\cos \theta). \quad (10)$$

We thereby obtain

$$\int_{-1}^{+1} RW(\theta, \tau) P_n(\cos \theta) d \cos \theta = e^{-n(n+1)\tau}, \quad (11)$$

or

$$\langle P_n(\cos \theta) \rangle_{RW} = e^{-n(n+1)\tau}, \quad (12)$$

where $\langle \rangle_{RW}$ denotes an average with respect to the random walk function. We now eliminate the parameter τ by observing that

$$J_s(T) = J_s(0) \langle P_1(\cos \theta) \rangle_{RW}, \quad (13)$$

which equation, combined with (12) leads to

$$\langle P_n(\cos \theta) \rangle_{RW} = \{J_s(T)/J_s(0)\}^{n(n+1)/2}. \quad (14)$$

Upon combining Eqs. (8) and (14), we obtain our final equation,

$$\langle S_n(\alpha_1, \alpha_2, \alpha_3) \rangle_{RW} = \{J_s(T)/J_s(0)\}^{n(n+1)/2} S_n(\bar{\alpha}_1, \bar{\alpha}_2, \bar{\alpha}_3). \quad (15)$$

Angular Distribution of 12- and 16-Mev Gamma Rays from the Proton Bombardment of a Thin Boron Target*

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The angular distribution of 12- and 16-Mev gamma rays from proton bombardment of thin boron targets has been obtained for proton energies ranging from 0.6 to 2.0 Mev. The angular distribution may be represented by the equation $w(\theta) = 1 + A \cos \theta + B \cos^2 \theta$, indicating interference between at least two levels of opposite parity. An analysis of the energy dependence of the coefficients A and B for the 16-Mev gamma rays indicates the interference to be between more than two levels.

INTRODUCTION

GAMMA-RAY resonances in the yield from the proton bombardment of boron have been observed by various investigators at 163,¹ 680,^{2,3} 1388, 2650,⁴ and 3550 keV,⁵ all except the first having very large widths. The excited states of C¹² may decay directly to the ground state with the emission of gamma rays of energy equal to the excitation energy ($Q=15.949$ Mev) or to the 4.43-Mev excited state and then to the ground state with the emission of cascade radiation. These gamma-rays are called, in the following, the "16-Mev," "12-Mev," and "4.43-Mev" radiations.

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¹ R. Tangen, Kgl. Norske. Videnskabs. Selskabs, Skriffter No. 1 (1946).

² Cochran, Ryan, Givin, Kern, and Hahn, Phys. Rev. **87**, 672 (1952).

³ T. Huus and R. Day, Phys. Rev. **91**, 599 (1953).

⁴ H. E. Gove and E. B. Paul, Phys. Rev. **91**, 463 (A) (1953).

⁵ H. B. Willard (private communication).

Earlier work⁶ has shown a $\cos \theta$ term in the angular distribution of the gamma radiation from this reaction over the energy region from 300 to 1100 keV. This indicates interference between at least two states of opposite parity. Since the spin and parity are known for the C¹² level corresponding to the 163-keV resonance, this investigation of the angular distribution of the 16-Mev gamma rays was undertaken so that the spins and parities of the interfering levels could be determined. Although the theoretical analysis of the angular distribution of the 12-Mev gamma rays is more complex, the experimental determination of this angular distribution was also undertaken since the spin and parity of the 4.43-Mev excited state of C¹² are known.

EXPERIMENTAL PROCEDURE

Protons were accelerated in the University of Kentucky 3-Mev electrostatic accelerator to energies

⁶ Jenkins, Cochran, Kern, and Hahn, Phys. Rev. **91**, 915 (1953).