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Theory of Dipole Interaction in Crystals *

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It is shown that dipole arrays may be represented as vectors in a many-dimensional vector space. The classical dipole interaction energy is a quadratic form in the components of the dipole moments. Its calculation is reduced to the diagonalization of this form. The characteristic vectors are so called basic arrays. An arbitrary array may be decomposed into a linear combination of basic arrays, the energies are additive and may be obtained from the characteristic values of the quadratic form. The method is demonstrated by the complete solution of the characteristic value problem of a highly symmetric class of cubic arrays. The minimum energy arrays are obtained without and with an external magnetic field for the simple cubic, body-centered cubic, and face-centered cubic lattices. The results are in good qualitative agreement with the experiments of de Haas and Wiersma on Cs Ti alum. Some discrepancies are attributed to quantum effects and to incomplete saturation (entropy $S>0$). The extension to these more general cases will be considered in a following paper.

I. INTRODUCTION

VARIOUS problems in the theory of solids lead to the consideration of interaction among dipoles. Typical examples are the dielectric and thermal behavior of certain crystals containing polar molecules and of most of the substances used in experiments on adiabatic demagnetization.

The paramagnetic substances which are suitable for these experiments contain magnetic moments whose freedom of orientation is restricted by the weakest possible interaction. This requirement is satisfied by magnetic ions containing an odd number of electrons. In fact, a theorem of Kramers[1] states that magnetic ions consisting of an odd number of electrons maintain a double degeneracy in any electrostatic field, and therefore the usually important crystalline Stark effect is ineffective in this case. However, Nernst's theorem requires that some mechanism should exist for removing this degeneracy and the splitting actually is a result only of the weaker forces from the direct coupling between spins. This coupling arises from the magnetic (dipole) forces, or to some extent from exchange forces. The dipole coupling is certainly more important. Moreover, in contrast to most types of interaction, it contains no unknown constants relating to atomic or crystalline structure. Thus the calculation of the magnetic interaction energies and of the partition sum is of added interest.

The present state of the theory of the dipole interaction is rather unsatisfactory. The simplest approach is the Lorentz field method whose shortcomings are well known.[2] That the magnetization is not the only factor in determining the internal field is most clearly visible from the calculations of Sauer[3] who computed the energies of certain intuitively selected dipole arrays and found that ordered arrays of zero magnetization may have widely different energies, some of them lower than that due to the Lorentz field. Sauer's calculation is valid only for absolute zero and vanishing external field.

A consistent theory has been developed by Van Vleck[2] who expands the partition sum of the crystal in decreasing powers of the temperature. This calculation fails at low temperatures where the interaction becomes really important.

Other attempts have been made to develop a theory for all values of the field and temperature[4] with no satisfactory results. The reason seems to be that all attempts were based on some kind of "nearest neighbor" method. However, the

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[1] H. A. Kramers, Proc. Amsterdam Acad. **33**, 959 (1930).
[2] Cf. J. H. Van Vleck, J. Chem. Phys. **5**, 320 (1937) and Ann. N. Y. Acad. Sci. **40**, 293 (1940).
[3] J. A. Sauer, Phys. Rev. **57**, 142 (1940).
[4] E.g. J. A. Sauer—A. N. V. Temperley, Proc. Roy. Soc. **176**, 203 (1940).

dipole interaction lends itself particularly badly to such kind of treatment. In the first place, the forces are of comparatively long range and even more important, their peculiar directional dependence makes the averaging over the directions inadequate.

The main point of the present paper is the use of a simple and rigorous "normal coordinate" method rather than the clearly inadequate "nearest neighbor" treatment of dipole interactions.

It will be shown in Section II that dipole arrays can be represented as vectors in a many dimensional space $\mathfrak{R}$. The dipole interaction energy appears as a quadratic form in the vector components of the dipole moments.

Thus the whole calculation is reduced to a characteristic value problem, the solution of which is greatly facilitated if the symmetry of the problem allows the application of group theory. The characteristic vectors will be called basic arrays (B.A.), their energies are the characteristic values of the quadratic form. The energy of any arbitrary array can be obtained by means of decomposition into a linear combination of basic arrays, as the energies of the arrays are additive. The arrays of minimum energy may also be obtained in a systematic manner.

The method is demonstrated by the complete solution of the problem for a very symmetrical class of simple cubic (S.C.) arrays (Section II). The results are extended to the body-centered (B.C.) and face-centered (F.C.) cubic arrays in Section IV. The numerical calculations are indicated and their results tabulated in Section III.

As the class of arrays considered in this paper is highly ordered, the results can be applied for real crystals only in case of vanishing entropy. These conditions are approximately realized for demagnetization experiments from high initial fields. Comparison with experiment is discussed in VI. Quantum effects and the extension to more general arrays will be discussed in a following paper.

II. VECTOR SPACE REPRESENTATION OF DIPOLE ARRAYS

Let us first consider S.C. dipole arrays obtained from S.C. lattices by placing a dipole of definite moment and direction at every lattice point (l.p.).

Dimensionless quantities will be used throughout this paper. Dipole moments will be measured in terms of an arbitrary dipole moment μ, length in terms of the lattice constant a. All magnetic (or electric) fields will be expressed in units of μ/a^3 and energies per unit volume in terms of $N^2\mu^2$ where N is the number of dipoles per unit volume. In the S.C., B.C., and F.C. cases one has $N=1/a^3$, $2/a^3$, $4/a^3$, respectively.

As indicated in the introduction, only ordered arrays will be considered in the present paper. A precise definition of an "ordered array" can be given by making use of the symmetry of the array. Let Γ be the group of cubic translations $l_1\mathbf{i}+l_2\mathbf{j}+l_3\mathbf{k}$ (l_1, l_2, l_3 are integers, $\mathbf{i}$, $\mathbf{j}$, $\mathbf{k}$, are unit vectors in the x, y, z direction).

The most completely ordered array is invariant under the same group, i.e., all its dipoles are equal and parallel. This array is of importance in building up others and will be called an S array. The dipole interaction energy of an S array is $-\frac{1}{2}(4\pi/3-l)$ where l is the demagnetization coefficient. For a spherical sample $l=4\pi/3$ and the energy vanishes.

A more general class of ordered arrays is obtained if invariance is required only under the subgroup Γ^2 of Γ consisting of the translations of the form $l_1(2\mathbf{i})+l_2(2\mathbf{j})+l_3(2\mathbf{k})$. Such arrays are said to be of the class Γ^2 and will be the only ones considered in this paper.[5]

To generate such arrays we have to specify 8 dipoles $\mathbf{p}^\nu$ ($\nu=1, 2, \cdots 8$), where ν is associated in some definite manner with corners of the unit cube having the coordinates $l_1, l_2, l_3=0, 1$. The whole array is constructed by the translations Γ^2.

The resulting array may be considered as a superposition of eight arrays each of which consists of parallel dipoles. These arrays are geometrically similar to the S arrays previously introduced, but have a lattice constant two (in units of a). These shall also be called S arrays;

[5] There are very good reasons to believe that the configurations of lowest (and highest) energy are of the class Γ^2. All arrays calculated previously[3] are of this class. No rigorous proof of this statement seems to be possible, however, before the investigation of the more general arrays has been carried out. We hope to come back to this question in a sequel to this paper.

in case of ambiguity the lattice constant will be specified. This point of view will be useful for the numerical calculations of Section III.

It is seen that every array of class Γ^2 can be specified by a set of 24 numbers, e.g., the three rectangular components of the 8 dipole moments p_x^ν, p_y^ν, $p_z^\nu = 1, 2, \cdots 8$. Also, in a more concise notation p_i, $i = 1, 2, \cdots 24$.

In the cases of practical interest the dipoles placed at the 8 cube corners will have moments of the same absolute value which will be denoted by p. For such an array the 24 numbers satisfy the 8 conditions:

$$(p_x^\nu)^2+(p_y^\nu)^2+(p_z^\nu)^2=p^2, \quad \nu=1, 2, \cdots 8. \quad (1)$$

It will, however, prove advantageous to temporarily disregard these conditions and admit arrays of unequal dipole moments into the class Γ^2. In this case every set of 24 real numbers defines a Γ^2 array and there is a one to one correspondence between these arrays and the points of a 24-dimensional vector space $\mathfrak{R}$. The arrays satisfying the conditions (1) will be called arrays of constant (dipole) strength p.[6] The corresponding points form a 16-dimensional hypersurface in $\mathfrak{R}$. This will be frequently used in what follows and will be briefly called the "constant dipole surface."

The operations of addition, multiplication with a scalar and taking the scalar product are defined in the usual manner.[7]

$$\text{(a)} \quad \mathbf{P}+\mathbf{Q}=\{\mathbf{p}^\nu+\mathbf{q}^\nu\},$$

$$\text{(b)} \quad c\mathbf{P}=\{c\mathbf{p}^\nu\}, \quad (2)$$

$$\text{(c)} \quad \mathbf{P}\cdot\mathbf{Q}=\sum_{\nu=1}^{8}\mathbf{p}^\nu\cdot\mathbf{q}^\nu.$$

The square of the *norm* of an array $\mathbf{P}$ is defined as

$$\mathbf{P}\cdot\mathbf{P}=\sum_{\nu=1}^{8}(\mathbf{p}^\nu)^2. \quad (3)$$

If the array $\mathbf{P}$ is of constant strength p, its norm is $8^{\frac{1}{2}}p$.

In order to compute the energy of an array $\mathbf{P}$ it is necessary to know the field generated by $\mathbf{P}$ at all the lattice points. Obviously, the field will have the same symmetry (Γ^2) as the array. Hence the set of vectors representing the field *at the lattice points* will again correspond to a vector in the space $\mathfrak{R}$, and will be denoted by $\mathbf{F}$.

The operation leading from any array $\mathbf{P}$ to its field $\mathbf{F}$ can be regarded as a mapping of the space $\mathfrak{R}$ on itself. One may write symbolically

$$\mathbf{F}=\mathfrak{F}\mathbf{P} \quad (4)$$

where $\mathfrak{F}$ is the "field operator." It is linear, as follows at once from the well-known expression for the field $\mathbf{f}$ of a dipole $\mathbf{p}$ at a point $\mathbf{r}$:

$$\mathbf{f}=(3\mathbf{r}(\mathbf{p}\cdot\mathbf{r})-\mathbf{p}r^2)/r^5. \quad (5)$$

The dipole interaction energy per unit volume is[8]

$$U=-(1/16)\mathbf{P}\cdot\mathbf{F}=-(1/16)\mathbf{P}\cdot\mathfrak{F}\mathbf{P}. \quad (6)$$

Equation (6) is an invariant relation independent of the choice of coordinate system. If, as above, we choose a coordinate system in which the array is represented by the rectangular components of the 8 dipole moments then one may rewrite Eq. (6) in matrix form:

$$U=-(1/16)\sum_{\mu,\nu=1}^{8}\sum_{x,y}\mathfrak{F}_{\mu\nu}{}^{xy}p_x p_y. \quad (7)$$

The matrix $\mathfrak{F}_{\mu\nu}{}^{xy}$ satisfies the symmetry relation

$$\mathfrak{F}_{\mu\nu}{}^{xy}=\mathfrak{F}_{\nu\mu}{}^{yx}. \quad (8)$$

This is a direct consequence of the existence of a potential energy for two dipoles; the energy can be considered as scalar product of the first dipole moment with the field due to the second, or *vice versa*.

It is sometimes convenient to write (7) and (8) in a more concise form by replacing the index couple μ, x by a single index i running from 1 to 24.

One has[9]

$$U=-(1/16)\sum_{i,j=1}^{24}\mathfrak{F}_{ij}p^i p^j. \quad (7a)$$

[6] The dipole strength of our array should be distinguished from its resultant dipole moment. The latter is proportional to the vector sum of the moments of the 8 cube corners.

[7] Boldfaced small letters will denote ordinary 3-dimensional vectors and boldfaced capital letters vectors in the 24-dimensional vector space.

[8] The numerical factor in this expression is explained as follows: the energy per unit volume is in our units the energy of one dipole, while (6) involves 8 dipoles. The additional factor $\frac{1}{2}$ corrects in the usual manner the fact that the interaction of every pair of dipoles is counted twice.

[9] In case of vectors in the space $\mathfrak{R}$ superscripts describe components and subscripts distinguish between different vectors.

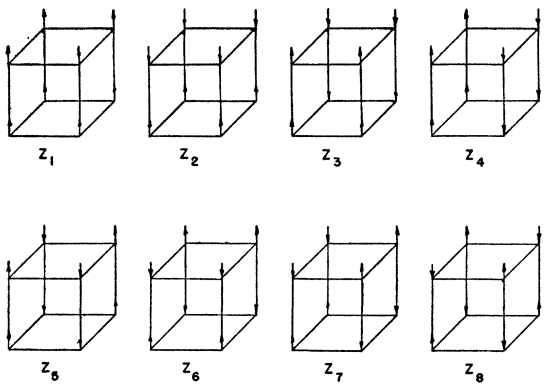

FIG. 1. The eight basic arrays. Interchange the labels Z_3 and Z_4.

The quadratic form (7) can be transformed into a sum of squares by means of an orthogonal transformation of the coordinate system in $\mathfrak{R}$ which leaves (3) invariant (rotation). The new coordinate system will be given by an orthogonal set of vectors $\mathbf{A}_i$, $i=1, 2, \cdots 24$, which will be called basic arrays (B.A.). These are closely analogous to the normal coordinates introduced for the description of vibrating systems. The calculation of the energy of an arbitrary array can then be reduced to finding the characteristic values of the operator $\mathfrak{F}$. This problem is greatly facilitated by group theoretical methods, based on the remark that the operator $\mathfrak{F}$ is invariant under the group $\mathbf{\Gamma}$.[10]

$$\Gamma\mathfrak{F}=\mathfrak{F}\Gamma. \tag{9}$$

This relation is intuitively evident, as it is immaterial whether a translation Γ is carried out on a field $\mathbf{F}$, or on a corresponding array $\mathbf{P}$ and the mapping leading to the field is carried out afterwards.

It follows from (9) by standard methods used in case of other linear operators (Schroedinger operator, vibrating systems) that the eigenvectors can be so chosen as to transform according to irreducible representations of the group $\mathbf{\Gamma}$.[11]

It may be remarked that so far no essential use has been made of the fact that the arrays $\mathbf{P}$ are of the class $\mathbf{\Gamma}^2$. If instead of $\mathbf{\Gamma}^2$ another sub-group of $\mathbf{\Gamma}$ had been chosen, the only difference in the above considerations would be that the number of dimensions of the space $\mathfrak{R}$ would be larger than 24. The formulae (1)–(9) will form the basis for a second paper, in which more general arrays will be discussed.

The actual solution of the eigenvalue problem is considerably simpler, however, for the class $\mathbf{\Gamma}^2$ than for the general classes.

A $\mathbf{\Gamma}^2$ array consisting of dipoles all pointing in, say, the x direction gives rise to fields at the l.p. pointing in the same direction, i.e.,

$$\mathfrak{F}_{\mu\nu}{}^{xy}=0 \text{ unless } x=y. \tag{10}$$

This is caused by the fact that a $\mathbf{\Gamma}^2$ array is invariant under a mirroring $y\rightarrow -y$, while the expression of the y component of the field consists of terms proportional to xy and thus changes sign.

In addition, because of the cubic rotational symmetry

$$\mathfrak{F}_{\mu\nu}{}^{xx}=\mathfrak{F}_{\mu\nu}{}^{yy}=\mathfrak{F}_{\mu\nu}{}^{zz}. \tag{11}$$

Thus the 24-dimensional matrix is reduced to three identical 8×8 matrices.

It is well known that the representations of the group $\mathbf{\Gamma}$ are the roots of unity. In the case of $\mathbf{\Gamma}^2$ arrays the relevant roots are the square roots $+1$ and -1. One is thus led uniquely to a definition of the B.A. which will be given now. The fact that they are B.A., i.e., characteristic vectors of the operator $\mathfrak{F}$ can be easily verified without any reference to group theory.

Corresponding to the reduction of the matrix $\mathfrak{F}$ into 3 identical 8 row matrices, the 24, B.A. fall into 3 groups, $\mathbf{X}_i$, $\mathbf{Y}_i$, $\mathbf{Z}_i$, $i=1, 2, \cdots 8$ consisting of dipoles pointing in the x, y, z directions, respectively.

The 8 non-vanishing components of the $\mathbf{Z}_i$ arrays are given by

$$Z_i=(-1)_{\alpha_i l_1+\beta_i l_2+\gamma_i l_3}, \quad i=1, 2, \cdots 8, \tag{12}$$

where α_i, β_i, $\gamma_i=0$, 1. It may be recalled that the superscripts are associated with the 8 cube corners l_1, l_2, $l_3=0$, 1. One has the following 8 possibilities:

	α	β	γ		α	β	γ
$\mathbf{Z}_1$	0	0	0	$\mathbf{Z}_5$	1	1	0
$\mathbf{Z}_2$	0	0	1	$\mathbf{Z}_6$	0	1	1
$\mathbf{Z}_3$	1	0	0	$\mathbf{Z}_7$	1	0	1
$\mathbf{Z}_4$	0	1	0	$\mathbf{Z}_8$	1	1	1

[10] In fact, it is invariant also under the group including the cubic rotations. However, this will be of no importance in the special case considered in the present paper.

[11] Cf. for instance B. L. van der Waerden, *Die Gruppentheoretische Methode in der Quanten Mechanik* (Edwards Brothers, New York, 1944). E. Wigner, Goettinger Nachrichten, p. 133 (1930).

These arrays are explicitly given in Fig. 1. $\mathbf{X}_i$ and $\mathbf{Y}_i$ are obtained from $\mathbf{Z}_i$ through cubic rotations, so that identical subscripts refer to identical geometric arrangements. Being basic arrays the $\mathbf{Z}_1 \cdots \mathbf{Z}_8$ are orthogonal. They obey the relations:

$$\mathbf{Z}_i \cdot \mathbf{Z}_j = 8\delta_{ij}, \tag{13}$$

and similarly the $\mathbf{X}_i$ and $\mathbf{Y}_i$.

Equation (13) can be verified either by means of Fig. 1 or algebraically as follows:

$$\mathbf{Z}_i \cdot \mathbf{Z}_j = \sum_{\nu=1}^{8} Z_i^{\nu} Z_j^{\nu}$$

$$= \sum \{l_1, l_2, l_3 = 0, 1\} (-)^{(\alpha_i+\alpha_j) l_1 + (\beta_i+\beta_j) l_2 + (\gamma_i+\gamma_j) l_3}.$$

If $i \neq j$, then at least one of the inequalities holds: $\alpha_i \neq \alpha_j$, $\beta_i \neq \beta_j$, $\gamma_i \neq \gamma_j$. Say $\alpha_i \neq \alpha_j$, then $\alpha_i + \alpha_j = 1$ and $\mathbf{Z}_i \cdot \mathbf{Z}_j = 0$, because of the summation over l_1.

$\mathbf{X}_i$, $\mathbf{Y}_i$, $\mathbf{Z}_i$ form a complete set of 24 orthogonal vectors and are the only ones which have the correct transformation properties. Hence they solve the eigenvalue problem:[12]

$$\mathfrak{F}\mathbf{X}_i = f_i \mathbf{X}_i, \quad \mathfrak{F}\mathbf{Y}_i = f_i \mathbf{Y}_i, \quad \mathfrak{F}\mathbf{Z}_i = f_i \mathbf{Z}_i. \tag{14}$$

Because of the completeness, every Γ^2 array $\mathbf{P}$ can be represented as

$$\mathbf{P} = \sum_{i=1}^{8} (a_i \mathbf{X}_i + b_i \mathbf{Y}_i + c_i \mathbf{Z}_i) \tag{15}$$

with $a_i = \frac{1}{8}\mathbf{P} \cdot \mathbf{X}_i$, $b_i = \frac{1}{8}\mathbf{P} \cdot \mathbf{Y}_i$, $c_i = \frac{1}{8}\mathbf{P} \cdot \mathbf{Z}_i$. The square of the norm of $\mathbf{P}$ is obtained in terms of the new coordinates from (3), (15) and the orthogonality relations:

$$\mathbf{P} \cdot \mathbf{P} = 8 \sum_{i=1}^{8} (a_i^2 + b_i^2 + c_i^2). \tag{16}$$

The factor 8 arises because the B.A. are normalized to have the dipole strength unity, and hence have the norm $8^{\frac{1}{2}}$. The field corresponding to $\mathbf{P}$ is

$$\mathbf{F} = \sum_{i=1}^{8} (a_i f_i \mathbf{X}_i + b_i f_i \mathbf{Y}_i + c_i f_i \mathbf{Z}_i), \tag{17}$$

and the energy per unit volume

$$U = -\tfrac{1}{2} \sum_{i=1}^{8} (a_i^2 + b_i^2 + c_i^2) f_i$$

$$= \sum_{i=1}^{8} (a_i^2 + b_i^2 + c_i^2) U_i, \tag{18}$$

where $U_i = -f_i/2$.

It is seen that the computation of the energy of any Γ^2 array is reduced to the knowledge of the characteristic values f_i. These have been computed by a method outlined in the next section and the values are to be found in Table II.

From the point of view of practical application there is also a somewhat different problem to be considered: given a S.C. crystal with dipoles of constant moment (taken as unity), but undetermined orientation, what is the Γ^2 array of lowest energy? Or, in the terminology introduced above: find the minimum value of the energy (18) for arrays lying on a given constant dipole surface. These arrays satisfy the 8 auxiliary conditions (1) which can be rewritten in terms of B.A. as follows:

$$\left(\sum_{i=1}^{8} a_i \mathbf{X}_i^{\nu}\right)^2 + \left(\sum_{i=1}^{8} b_i \mathbf{Y}_i^{\nu}\right)^2 + \left(\sum_{i=1}^{8} c_i \mathbf{Z}_i^{\nu}\right)^2 = 1, \tag{19}$$

$$\nu = 1, 2, \cdots 8.$$

The standard procedure accounting for the auxiliary conditions by the method of Lagrange multipliers proves to be very cumbersome. The following artifice yields the desired result without any further calculation, not only in the simple case considered here, but also in some of the more complicated cases discussed in Sections IV and V: The condition of constant dipole strength unity implies that the norm of the array is $8^{\frac{1}{2}}$, or, using (16),

$$\sum_{i=1}^{8} (a_i^2 + b_i^2 + c_i^2) = 1. \tag{20}$$

The conditions (19) imply (20), but not *vice versa*. Therefore they will be called briefly the strong and the weak conditions, respectively.

[12] Equation (14) could be easily verified directly using the explicit expressions of Section III.

The procedure consists in minimizing the energy (18) under the weak condition alone. This can be done at once by means of the well-known extremum property of the characteristic values.[13]

The lowest value of the energy is $-f_m/2$ where f_m is the greatest characteristic value of the operator $\mathfrak{F}$. The array is a linear combination of the B.A. corresponding to f_m. If some of these linear combinations satisfy the strong conditions, then the original problem is solved.

It is seen from Table II that the lowest energy for the S.C. lattice is $-f_5/2=-2.676$. The corresponding array is $a_5\mathbf{X}_5+b_5\mathbf{Y}_5+c_5\mathbf{Z}_5$ with $a_5^2+b_5^2+c_5^2=1$. It is easily seen that this array satisfies the strong condition and represents the correct solution of the problem.

III. CALCULATION OF THE FIELDS

The object of this section is the computation of the characteristic values f_i defined in the preceding section. According to its definition f is the value of the field of the ith B.A. say $\mathbf{Z}_i$ at a lattice point. It was pointed out at the beginning of Section II that any Γ^2 array (and thus in particular $\mathbf{Z}_i$) can be considered as a superposition of eight S arrays of lattice constant two. Hence, the field at any point of a B.A. will be known as soon as the field of an S array is known at every point. We shall denote the field of a z directed S array as[14] $\mathbf{S}(\mathbf{r})$, $\mathbf{r}$ being the location of the point in question.

Using the expression (5) for the field of a dipole we arrive at the following equations for the x, y, and z components

$$S_x(\mathbf{r})=\sum_{-\infty}^{\infty}{}_{\{l_1,\,l_2,\,l_3\}}\frac{3(l_1-x)(l_3-z)}{[(l_1-x)^2+(l_2-y)^2+(l_3-z)^2]^{5/2}},$$

$$S_y(\mathbf{r})=\sum_{-\infty}^{\infty}{}_{\{l_1,\,l_2,\,l_3\}}\frac{3(l_2-y)(l_3-z)}{[(l_1-x)^2+(l_2-y)^2+(l_3-z)^2]^{5/2}}, \tag{21}$$

$$S_z(\mathbf{r})=\sum_{-\infty}^{\infty}{}_{\{l_1,\,l_2,\,l_3\}}\frac{2(l_3-z)^2-(l_1-x)^2-(l_2-y)^2}{[(l_1-x)^2+(l_2-y)^2+(l_3-z)^2]^{5/2}}.$$

Using the function $\mathbf{S}(\mathbf{r})$ (which we shall call the "characteristic function"), we may write the field $\mathbf{H}_i$ at any point $\mathbf{r}$ of $\mathbf{Z}_i$ explicitly as

$$\mathbf{H}_i(\mathbf{r})=\tfrac{1}{8}\sum_{\{l_1,\,l_2,\,l_3=0,\,1\}}(-)^{\alpha_i l_1+\beta_i l_2+\gamma_i l_3}\times\mathbf{S}\left(\frac{l_1-x}{2},\frac{l_2-y}{2},\frac{l_3-z}{2}\right), \tag{22}$$

where α_i, β_i, γ_i correspond to the B.A. $\mathbf{Z}_i$. The $\frac{1}{2}$ in the argument of the characteristic function and the $\frac{1}{8}$ multiplying the entire expression arise from the fact that the characteristic function is defined for an S array with lattice constant unity, while the component S arrays of a B.A. have lattice constant equal to two. Thus the question of finding the characteristic values is reduced to the knowledge of the values of the function S at the points with coordinates having half integral values.

It will be seen in the next section that the solution of the characteristic value problem for the B.C. and F.C. lattices necessitates the knowledge of the field in the body centers and face centers. These may also be obtained provided a few more values of the function $\mathbf{S}(\mathbf{r})$ are computed.

In the actual computation of the fields full use is made of symmetry considerations, which show that at many points the field is zero, or is simply related to the field at other points. As a typical example, we shall show that the field of $\mathbf{Z}_6$ at a lattice point is minus one-half the field of $\mathbf{Z}_5$ at a lattice point. From the expression for the dipole interaction (5) and the definition of the

[13] C.f. for example, Courant-Hilbert, *Methoden d. Mathematischen Physik* (Verlagsbuchhandlung Julius Springer, Berlin, 1931), Vol. I, p. 26.

[14] $\mathbf{S}(\mathbf{r})$ is a spatial vector point function of the points r within the unit cell, and should not be confused with the 24 dimensional vectors representing the field *at the lattice points*.

TABLE I. Values of the fields. The values given for Z_1 are valid for spherical samples. Otherwise $((4\pi/3)-l)Z_1$ should be added to every term in the first line. l is the demagnetization coefficient. The numerical values of f_i, g, h_i are to be found in Table II.

Array	Field at l.p.	Field at B.C.	Fields at F.C. XY Face	YZ Face	ZX Face
$\mathbf{Z}_1$	0	0	$-2h_1\mathbf{Z}_1$	$h_1\mathbf{Z}_1$	$h_1\mathbf{Z}_1$
$\mathbf{Z}_2$	$f_2\mathbf{Z}_2$	0	$-2h_2\mathbf{Z}_2$	0	0
$\mathbf{Z}_3$	$f_3\mathbf{Z}_3$	0	0	$h_2\mathbf{Z}_3$	0
$\mathbf{Z}_4$	$f_4\mathbf{Z}_4$	0	0	0	$h_2\mathbf{Z}_4$
$\mathbf{Z}_5$	$f_3\mathbf{Z}_5$	0	0	0	0
$\mathbf{Z}_6$	$f_6\mathbf{Z}_6$	$g\mathbf{Y}_7$	0	$h_3\mathbf{Y}_7$	0
$\mathbf{Z}_7$	$f_7\mathbf{Z}_7$	$g\mathbf{X}_6$	0	0	$h_3\mathbf{X}_6$
$\mathbf{Z}_8$	0	0	0	$h_4\mathbf{Y}_8$	$h_4\mathbf{X}_8$

basic arrays one easily finds

$$H_{5z}(0,0,0)=\sum{}'\frac{2l_3^2-l_1^2-l_2^2}{(l_1^2+l_2^2+l_3^2)^{5/2}}(-)^{l_1+l_2}, \quad (23)$$

$$H_{6z}(0,0,0)=\sum{}'\frac{2l_3^2-l_1^2-l_2^2}{(l_1^2+l_2^2+l_3^2)^{5/2}}(-)^{l_2+l_3}; \quad (24)$$

noticing that l_1 and l_2 enter into (23) in the same manner, we get

$$H_{5z}=2\sum{}'\frac{l_3^2-l_2^2}{(l_1^2+l_2^2+l_3^2)^{5/2}}(-)^{l_1+l_2}.$$

Interchanging[15] l_1 and l_3 in (24) we get

$$H_{6z}=\sum{}'\frac{2l_1^2-l_3^2-l_2^2}{(l_1^2+l_2^2+l_3^2)^{5/2}}(-)^{l_1+l_2}$$

$$=\sum{}'\frac{l_2^2-l_3^2}{l_1^2+l_2^2+l_3^2}(-)^{l_1+l_2}=-\tfrac{1}{2}H_{5z}$$

which is the required result.

Many other relationships exist, connecting the different fields at the body centers with each other, connecting different lattice point fields, different face-center fields, etc. By means of these relationships it is possible to calculate the fields at the lattice points, body centers, and face centers of a B.A. by computing only six different values of the characteristic function $\mathbf{S}(\mathbf{r})$. These are $S_z(\frac{1}{2},0,0)$, $S_z(0,\frac{1}{2},\frac{1}{2})$, $S_y(\frac{1}{4},\frac{1}{4},\frac{1}{4})$, $S_z(0,\frac{1}{4},\frac{1}{4})$, $S_y(0,\frac{1}{4},\frac{1}{4})$, $S_y(\frac{1}{2},\frac{1}{4},\frac{1}{4})$. It is clear that others could have been chosen, but these turn out to be convenient. Table I gives all the fields expressed in terms of these fields. The numerical values of the first three have been taken from a paper of McKeehan,[16] while the others have been calculated using the Ewald[17] method. A check on our values may be obtained from McKeehan since it is possible to evaluate $S_y(0,\frac{1}{4},\frac{1}{4})-S_y(\frac{1}{2},\frac{1}{4},\frac{1}{4})$ from his tables. The agreement is excellent. The values of these fields are:

$$S_z(\tfrac{1}{2},0,0)=15.040, \quad S_z(0,\tfrac{1}{4},\tfrac{1}{4})=31.521,$$
$$S_z(0,\tfrac{1}{2},1)=4.334, \quad S_y(0,\tfrac{1}{4},\tfrac{1}{4})=2.599,$$
$$S_y(\tfrac{1}{4},\tfrac{1}{4},\tfrac{1}{4})=10.620, \quad S_y(\tfrac{1}{2},\tfrac{1}{4},\tfrac{1}{4})=12.329.$$

Tables I and II give the resulting values of all the fields.

TABLE II. Characteristic values f, g, and h.

$f_2=$	$\frac{1}{2}[S_z(0,\frac{1}{2},\frac{1}{2})-S_z(\frac{1}{2},0,0)]=-9.687$
$f_3=$	$-\frac{1}{4}[S_z(0,\frac{1}{2},\frac{1}{2})-S_z(\frac{1}{2},0,0)]=4.844$
$f_4=$	$-\frac{1}{4}[S_z(0,\frac{1}{2},\frac{1}{2})-S_z(\frac{1}{2},0,0)]=4.844$
$f_5=$	$-\frac{1}{2}[S_z(0,\frac{1}{2},\frac{1}{2})+S_z(\frac{1}{2},0,0)]=5.351$
$f_6=$	$\frac{1}{4}[S_z(0,\frac{1}{2},\frac{1}{2})+S_z(\frac{1}{2},0,0)]=-2.676$
$f_7=$	$\frac{1}{4}[S_z(0,\frac{1}{2},\frac{1}{2})+S_z(\frac{1}{2},0,0)]=-2.676$
$g=$	$S_y(\frac{1}{4},\frac{1}{4},\frac{1}{4})=10.620$
$h_1=$	$S_z(0,\frac{1}{2},\frac{1}{2})=4.334$
$h_2=$	$S_z(0,\frac{1}{4},\frac{1}{4})-S_z(0,\frac{1}{2},\frac{1}{2})=7.992$
$h_3=$	$\frac{1}{2}[S_y(0,\frac{1}{4},\frac{1}{4})+S_y(\frac{1}{2},\frac{1}{4},\frac{1}{4})]=17.065$
$h_4=$	$\frac{1}{2}[S_y(0,\frac{1}{4},\frac{1}{4})-S_y(\frac{1}{2},\frac{1}{4},\frac{1}{4})]=14.461$

IV. BODY-CENTERED AND FACE-CENTERED ARRAYS

It is convenient to consider the B.C. and F.C. arrays as consisting of two and four S.C. arrays, respectively, which can be resolved into B.A. In this representation the field matrix contains diagonal elements corresponding to the energies of the constituent S.C. arrays and off-diagonal elements giving the interaction of B.A. at different points. The interaction terms are listed in Tables I and II. It is apparent that most of the off-diagonal terms vanish and the energy of any B.C. or F.C. array can be readily computed. As an example, we have considered a set of

[15] Such procedures may be justified by transforming these series to absolutely convergent ones by means of the Ewald method. For details see J. Bouman, Archives Neerlandaises [3A], **13**, 1–28 (1931).

[16] L. W. McKeehan, Phys. Rev. **43**, 913 (1933). McKeehan gives values to five decimal places. However, by means of our symmetry relations we can check some of his values. While the agreement is generally very good, sometimes it does not extend beyond the third decimal place. The uncertain digits have been omitted.

[17] P. P. Ewald, Ann. d. Physik **64**, 253 (1921). See also H. Mueller, Phys. Rev. **50**, 547 (1936); M. Born and H. Kornfeld, Physik. Zeits. **24**, 121 (1923); and H. Kornfeld, Zeits. f. Physik **22**, 27 (1924).

arrays previously computed by Sauer.[3] Table III compares the energies resulting from the decomposition into B.A. with the values obtained by Sauer through direct summation.

This representation does not lead in any systematic way to the minimum energy configuration of an array of given dipole strength. The latter problem can be solved by completing the diagonalization of the field matrix and introducing B.A. in the 48- and 96-dimensional vector spaces corresponding to B.C. and F.C. arrays, respectively. Since most of the off-diagonal terms vanished in the above representation, this can be easily carried out. The procedure will be explained in detail for the B.C. case.

It is seen from Table II that if one of the B.A. $\mathbf{X}_i$, $\mathbf{Y}_i$, $\mathbf{Z}_i$ ($i \neq 6, 7$) is placed at the lattice points it gives rise to no field at the body center. Similarly, one of these arrays placed at the body centers will give rise to no field at the lattice points, as the lattice points and body centers are inter-changeable. Thus by placing $\mathbf{X}_i$ at the lattice points and nothing at the body centers, we obtain a B.A. and similarly placing $\mathbf{X}_i$ at the body centers and nothing at the lattice points will also give B.A. We now introduce the notation $[\mathbf{P}, \mathbf{Q}]$ to denote a B.C. array with $\mathbf{P}$ at the lattice points and $\mathbf{Q}$ at the body centers. The above B.A. will then be written as $[\mathbf{X}_i, \mathbf{O}]$ and $[\mathbf{O}, \mathbf{X}_i]$, respectively. Using the same process on $\mathbf{Y}_i$, $\mathbf{Z}_i$, we obtain four more B.A. $[\mathbf{Y}_i, \mathbf{O}]$, $[\mathbf{O}, \mathbf{Y}_i]$, $[\mathbf{Z}_i, \mathbf{O}]$, $[\mathbf{O}, \mathbf{Z}_i]$. These six B.A. all correspond to the same characteristic value f_i of the field operators $\mathfrak{F}$, therefore any set of six orthogonal linear combinations of these B.A. will also be B.A. In view of the considerations at the end of II, it is convenient to choose B.A. having constant dipole strength. Such a choice would be $[\mathbf{Z}_i, \mathbf{Z}_i]$ and $[\mathbf{Z}_i, -\mathbf{Z}_i]$ and similarly for $\mathbf{X}_i$, $\mathbf{Y}_i$.

The number 6 and 7 arrays require special consideration as they give rise to off-diagonal terms in the field matrix. In other words, there is an interaction between, for example, a $\mathbf{Z}_6$ array at the lattice points and a $\mathbf{Y}_7$ array at the body centers. The diagonalization is easily completed by choosing $[\mathbf{Z}_6, \mathbf{Y}_7]$ and $[\mathbf{Z}_6, -\mathbf{Y}_7]$ as B.A. The corresponding characteristic values are f_5+g and f_6-g. Similar B.A. are constructed from the other 6 and 7 arrays.

These results are summed up in Table IV.

The F.C. case can be discussed in exactly the same manner. The treatment is somewhat more complicated because of the presence of many more interaction terms. Only the results are given (Table V). In order to obtain the energy values in units of $N^2\mu^2$ one has to multiply the characteristic values of $\mathfrak{F}$ by $(-\frac{1}{4})$ in the B.C. case and $(-\frac{1}{8})$ in the F.C. case.

Since all the B.A. are defined so as to have a constant dipole strength, the minimum energy configurations are simply obtained by choosing the highest characteristic values from Tables IV and V. Hence, the minimum energy for the B.C. case is $-\frac{1}{4}(g+f_6)N^2\mu^2 = 1.986N^2\mu^2$ and for the F.C. case $-(h_4/8)N^2\mu^2 = -1.808N^2\mu^2$.

It may be noted that Sauer correctly guessed one of the minimum energy arrays in the S.C. and F.C. case, but not in the B.C. case.

Finally, the possibility of "ferromagnetism" for these arrays should be discussed. Summing up our results, we notice that in all cases the minimum energy configuration has been non-

TABLE III. Arrays calculated by Sauer. (Sauer's symbols are in the first column.)

Type	Resolution into basic arrays	Energy constants: Sauer	Energy constants: Present paper
"A"			
a	Z_5	−2.7	−2.676
b	Z_1 at l.p., $-Z_1$ at b.c.	0	0
c	$Z_5+X_7+Y_6$ at l.p. and b.c.	−1.75	−1.770
d	Z_1 at l.p. and XY f.c. $-Z_1$ at YZ and ZX f.c.	2.2	2.167
e	Z_1+Y_1 at l.p. and YZ f.c. $-Z_1-Y_1$ at XY and XZ f.c.	−1.1	−1.084
"B"			
a	Z_5	−2.7	−2.676
b	Z_5 at l.p., $-Z_5$ at b.c.	−1.35	−1.338
c	$Z_5+X_7+Y_6$ at l.p. and b.c.	−1.75	−1.770
d	Z_1 at l.p. and YZ f.c. $-Z_1$ at XY and XZ f.c.	−1.1	−1.084
e	Z_8+Y_8 at l.p., XY and YZ f.c. $-Z_8-Y_8$ at ZX f.c.	−1.8	−1.808

a is S.C., dipole direction 001
b is B.C., dipole direction 001
c is B.C., dipole direction 111
d is F.C., dipole direction 001
e is F.C., dipole direction 011
"A" is an array which has nearest neighbor strings of antiparallel dipoles
"B" is an array which has nearest neighbor strings of antiparallel dipoles if the dipoles are contained in a plane perpendicular to the dipole direction, and passing through the dipole.

polarized. This result, however, is true only for spherical samples. Otherwise one has to add a term $-\frac{1}{2}(4\pi/3-l)N^2\mu^2$ to the energy of the polarized number 1 arrays, for all three cubic types. In the extreme case of a very long thin needle $l=0$, and the energy constant becomes $-2\pi/3=-2.094$, while the energy constants of the lowest non-polarized arrays are for S.C. -2.675, for B.C. -1.986, and for F.C. -1.808. Thus the S.C. array is always non-ferromagnetic, while the other cases should exhibit ferromagnetism for long thin needles. In the case of a F.C. lattice cut in the form of a prolate spheroid the ferromagnetic state is favored above an axis ratio of 6:1. This result has been found before by Sauer.[3] Whether this ferromagnetic state has a physical reality is, however, subject to some doubt. This question is taken up once more in VI.

V. DIPOLE ARRAYS IN A MAGNETIC FIELD

The energy of a given Γ^2 array in a magnetic field is easily calculated. Considering first the S.C. case, one has:

$$U=-\tfrac{1}{2}\sum_{i=1}^{8} f_i(a_i^2+b_i^2+c_i^2)-(a_1H_x+b_1H_y+C_1H_z) -(4\pi/3-l)(a_1^2+b_1^2+c_1^2)/2, \quad (25)$$

since only the number 1 arrays have a resultant magnetic moment. Let us introduce the notation $a_1^2+b_1^2+c_1^2=q^2$, q being the magnetization of the array in units of $N\mu$. Denoting the angle between magnetization and external field by ϑ (25) becomes

$$U=-\tfrac{1}{2}\textstyle\sum_i f_i(a_i^2+b_i^2+c_i^2) -qH\cos\vartheta-(4\pi/3-l)q^2/2. \quad (26)$$

TABLE IV. Characteristic values and typical basic arrays in the B.C. case. Valid for spherical sample, otherwise, cf. Table I.

Characteristic value of $\mathfrak{F}$	Degree of degeneracy	Typical B.A. At lattice points	At body centers
$f_1=f_8=$ 0	6	$\mathbf{Z}_1$	$\mathbf{Z}_1$
	6	$\mathbf{Z}_8$	$\mathbf{Z}_8$
$f_2=-$ 9.687	6	$\mathbf{Z}_2$	$\mathbf{Z}_2$
$f_3=f_4=$ 4.844	12	$\mathbf{Z}_3$	$\mathbf{Z}_3$
$f_5=$ 5.351	6	$\mathbf{Z}_5$	$\mathbf{Z}_5$
$f_6+g=$ 7.944	6	$\mathbf{Z}_6$	$\mathbf{Y}_7$
$f_6-g=-13.296$	6	$\mathbf{Z}_6$	$-\mathbf{Y}_7$

This expression should be minimized under the strong condition (19), which we again replace temporarily by the weak condition (20). In our present notation (20) takes the form:

$$\sum_{i=2}^{8}(a_i^2+b_i^2+c_i^2)+q^2=1 \quad (27)$$

We now minimize the energy at fixed q. From (26) we see that $\cos\vartheta=1$, i.e., the magnetization is parallel to the magnetic field. The minimization of the first term is exactly the problem solved at the end of II since $f_1=0$. The solution is

$$\mathbf{P}=a_1\mathbf{X}_1+b_1\mathbf{Y}_1+c_1\mathbf{Z}_1+a_5\mathbf{X}_5+b_5\mathbf{Y}_5+c_5\mathbf{Z}_5, \quad (28)$$

with $a_5^2+b_5^2+c_5^2=1-q^2$.

The energy becomes

$$U=-\tfrac{1}{2}(1-q^2)f_5-qH-(4\pi/3-l)q^2/2. \quad (29)$$

Equation (29) may now be minimized with respect to q: $\partial U/\partial q=0$ leads to

$$q=\frac{H}{f_5-(4\pi/3-l)}.$$

Defining a critical field H_c as

$$H_c=f_5-(4\pi/3-l) \quad (30)$$

and remembering that $q\leqq 1$, we have

$$q=\begin{cases} H/H_c & \text{for } H\leq H_c \\ 1 & \text{for } H\geq H_c. \end{cases} \quad (31)$$

In other words there exists a magnetic field H_c above which the magnetization is constant (saturation) and below which it drops to zero linearly with the field. It is recalled that the magnetization is given in units of $N\mu$.

Whether or not (28) is the correct solution is still dependent on whether it satisfies the strong conditions. This is generally not the case for an arbitrary direction of the magnetic field ($a_1\neq 0$, $b_1\neq 0$, $c_1\neq 0$). If, however, the magnetic field is along one of the cubic axis or in one of cubic planes, then the resulting array can be chosen to have constant dipole strength. For example, if the field is along the cubic axis, say in the Z direction, then the array

$$\mathbf{P}=a_5\mathbf{X}_5+b_5\mathbf{Y}_5+c_1\mathbf{Z}_1$$

satisfies all the requirements.

TABLE V. Characteristic values and typical basic arrays in the F.C. case. Valid for spherical samples, otherwise, cf. Table I.

Characteristic value of $\mathfrak{F}$	Degree of degeneracy	Typical B.A. with this characteristic value			
		At l.p.	At XY face	At YZ face	At ZX face
0	3	Z_1	Z_1	Z_1	Z_1
$-4h_1=-17.336$	3	Z_1	Z_1	$-Z_1$	$-Z_1$
$2h_1=8.668$	6	Z_1	$-Z_1$	Z_1	$-Z_1$
$f_2-2h_2=-25.671$	6	Z_2	Z_2	Z_2	$-Z_2$
$f_2+2h_2=6.297$	6	Z_2	$-Z_2$	Z_2	Z_2
$f_3+h_2=12.836$	12	Z_3	Z_3	Z_3	Z_3
$f_3-h_2=-3.148$	12	Z_3	Z_3	$-Z_3$	$-Z_3$
$f_5=5.351$	12	Z_5	Z_5	Z_5	Z_5
$f_6+h_3=14.389$	12	Z_6	Z_6	Y_7	$-Y_7$
$f_6-h_3=-19.741$	12	Z_6	Z_6	$-Y_7$	Y_7
$h_4=14.461$	4	Z_8	Z_8	Y_8	$-Y_8$
$h_4=14.461$	4	$2X_8+Y_8-Z_8$	$2X_8+Y_8-Z_8$	$2X_8+Y_8-Z_8$	$2X_8-Y_8+Z_8$
$-2h_4=-28.922$	4	$X_8-Y_8-Z_8$	$X_8-Y_8+Z_8$	$X_8+Y_8+Z_8$	$X_8+Y_8-Z_8$

Since in the general case the simple artifice of first ignoring the strong conditions does not work, one has to introduce (19) at the outset. This can be done by the method of Lagrange multipliers, but the resulting equations are very complicated and have not been solved. The case of physical interest is the F.C. array (paramagnetic alums), and here the simple method works once more.

The above considerations may be repeated for the F.C. case. Equations (25) through (31) are maintained provided the characteristic values and B.A. defined for the S.C. case for the space $\mathfrak{R}$ are replaced by the corresponding quantities in the 96-dimensional space. In particular the energy constant $-f_5/2$ should be replaced by $-h_4/8$. The minimum energy array under the weak condition is a superposition of the polarized B.A. and those belonging to the characteristic value h_4. Here, however, the formal identity ceases. Actually, the situation is more favorable than in the S.C. case as the strong conditions may now be satisfied for an arbitrary magnetic field. The array which satisfies the strong conditions is a superposition of the polarized arrays and those arrays belonging to the second class of h_4 arrays given in Table V. In the case $H_c \leq 0$ ferromagnetism will exist. Cf. IV.

VI. DISCUSSION

The main purpose of the present paper is to demonstrate the application of the proposed method and to lay the groundwork for generalizations. These would include, first of all, a treatment of the statistical mechanics of dipole arrays which requires the discussion of more general arrays than those of the class Γ^2, and a method of evaluating the partition sum in terms of them. The statistical problem must be solved before the theory may be applied to cases in which the entropy is finite (not equal to zero) such as is found for the hydrogen halide crystals in the neighborhood of their phase transition.

The ordered arrays of class Γ^2 should, however, be expected to give an adequate description of paramagnetic crystals containing ions with one valence electron (Ti, for example) in a state of zero entropy. Such a state is realized to a high degree of approximation for the entire process of adiabatic demagnetization with high initial fields. Although in reality the spin system should be treated quantum mechanically, it may be expected that the present classical method will give the essential features. A rigorous quantum treatment in which the Hamiltonian is set up in terms of the Pauli spin matrices is being planned for a sequel to this paper.

The computation of the magnetization curve in V will now be compared with the measurements of de Haas and Wiersma[18] on Cs–Ti alum. The salient feature of these experiments is that the magnetization stayed constant from $H=24{,}000$ gauss (approximately 90 percent saturation) to a field of about 100 gauss, and then dropped to zero almost linearly with the

[18] W. J. de Haas and E. C. Wiersma, Physica **3**, 491 (1936); cf. also H. B. O. Casimir, *Magnetism and Very Low Temperatures* (Cambridge University Press, England, 1940), p. 74.

field. This behavior is in excellent agreement with the results of Section V. The calculation leads to a critical field of $H_c = 65$ gauss which is the correct order of magnitude. The theory of this experiment has been worked out by Van Vleck[19] as an application of his general method.[2] The resulting magnetization curve is much too smooth. In fact, it is well known that no discontinuity of any kind can be expected on the basis of a "virial coefficients" method.

The magnetization could also be obtained on the basis of a theory of Sauer and Temperley.[4] This leads to a critical field below which the magnetization drops at once to zero. Such a result is an immediate consequence of their assumption that the spins are always parallel or antiparallel to the field. Hence, in this model there is no possibility of a state with vanishing entropy and having a magnetization intermediate between zero and the saturation value. This coupling scheme corresponds to that used in the case of the Paschen-Back effect. Actually for $H < H_c$ one is in the region of anomalous Zeeman effect. The sharpness of the transition arises from the fact that one is dealing with a crystal rather than an atom.

In the case of alums containing magnetic ions with more than one electron the magnetization curve is very much influenced by the crystalline field and the present theory may not be used alone. The dipole interaction, however, should be the decisive factor for the state of the system at absolute zero if the number of electrons in the magnetic ions is odd. The question of main interest is whether or not the state will be polarized (ferromagnetism or anti-ferromagnetism). In IV it was shown that for spherical samples the lowest state was always non-polarized, but that in the important F.C. case (to which the paramagnetic alums belong) a prolate spheroidal sample of axis ratio larger than 6, the polarized state will have the lower energy. This is of interest in connection the "ferromagnetism" observed by Kürti, Lainé, and Simon[20] for iron ammonium alum at low temperatures. That a stable ferromagnetic state really exists is still open to some question. In the first place, the numerical values of the energy constants might be changed by the above mentioned quantum mechanical treatment. In addition, it should be emphasized that pure energy considerations cannot completely decide questions of stability. The forces tending to polarize the crystal originate in distant parts of the sample, while the depolarization forces are due to relatively near neighbors. If the sample is cooled to a temperature below the Curie point by contact with a heat bath, rather than by adiabatic demagnetization, a ferromagnetic state would certainly not be established. This is not to be confused with the situation in true ferromagnetics, where the saturation moment is masked only by the domain structure. The question needs further study, both from the theoretical and the experimental point of view. It may certainly be said, however, that dipole ferromagnetism—if it exists—has a character essentially different from exchange ferromagnetism.

Finally, it may be noted that the more general developments of II are by no means restricted to cubic crystals. The difference in a non-cubic crystal would be in the characteric function $\mathbf{S}(\mathbf{r})$ defined in III, and in the detailed structure of the B.A. The calculations are much more difficult for the non-cubic case, however.

The method is not even restricted to dipole forces, but could be extended to Coulomb, quadrupole or exchange interactions. In fact, the method is applicable to any interaction which is quadratic in a property of the source.

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[19] J. H. Van Vleck, J. Chem. Phys. 6, 81 (1938).

[20] N. Kürti, P. Lainé, and F. Simon, Comptes rendus 204, 675 (1937).