# Antiferromagnetic Arrangements in Ferrites

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The molecular field treatment of the magnetic properties of ferrites given by Néel is extended to take into account the antiferromagnetic exchange interactions within the two magnetic sublattices. On further subdividing the two lattices, we show that the ground state may, (1) have an antiparallel arrangement of the spins on the two sites; or (2) consist of a triangular arrangement of the spins on the sublattices; or (3) have antiferromagnetism in each of the two sites separately. We also show that transitions between various configurations may occur in the same substance at different temperatures, without the assumption of temperature-dependent interactions. Neutron diffraction experiments and specific heat measurements are suggested for the detection of the predicted arrangements.

#### I. INTRODUCTION

W E are concerned in this paper with the theory of the magnetic properties of ferrites. We demonstrate in particular the possibility that both ferromagnetic and antiferromagnetic transitions, as well as transitions to triangular arrangements of the spins, may occur at different temperatures in the same substance.

Néel<sup>1</sup> has given a simple model which is known to account satisfactorily for many of the magnetic properties of the ferrites. We follow, and extend as required, his model and method of calculation, based on a Weiss molecular field treatment of the exchange interactions between the several spin lattices. On the Néel theory the magnetic ions are assumed to be distributed among the tetrahedral sites A and the octahedral sites B of the spinel structure.<sup>2</sup> Negative (antiferromagnetic) exchange interactions exist between A and A, A and B, and B and B.

When the A-B antiferromagnetic interaction is the dominant one, A and B will be magnetized in opposite directions below a transition temperature, giving rise, when the magnetizations of the A and B sites differ in magnitude, to a special case of ferromagnetism termed "ferrimagnetism." When the A - A (or B - B) interaction is dominant, Néel found that the above transition will not take place, and he concluded that the substance remains paramagnetic down to the lowest temperatures. This conclusion is not correct, as in the presence of strong interactions some kind of ordering may be expected to occur at low temperatures. An example of this case is provided by the mixed ferrite  $Fe_2O_3$ , xNiO, (1-x)ZnO. Pure Ni ferrite has the inverse spinel structure, and the A-B interaction is dominant. Pure Zn ferrite has the normal spinel structure;  $Zn^{++}$  being nonmagnetic, the A sites carry no magnetic moment. The B-B interaction is expected then to give rise to antiferromagnetic ordering in the B lattice. As x is varied from 0 to 1, intermediate arrangements will arise.

The purpose of the present paper is to study the intermediate arrangements and the associated Curie temperatures, extending Néel's scheme. We find several quite new effects, namely, that:

(a) The ground state at  $0^{\circ}$ K may have one of the following three configurations: (1) The spins on one site are antiparallel to those on the other (ferrimagnetism). (2) The spins on one site fall into two similar sublattices, each sublattice being ferromagnetically saturated. These magnetizations are at an angle to each other, and their resultant is antiparallel to the saturation magnetization of the spins on the other site (triangular case). (3) The spins on each of the two sites have antiferromagnetism.

(b) The transition temperature at which ordering first appears on cooling down the substance is not always such as to lead to the lowest configuration at  $0^{\circ}$ K (this is in contrast to the situation in simple antiferromagnetics where the highest Curie point is always such as to lead to the lowest state at  $0^{\circ}$ K), so that even with temperature-independent interaction coefficients the substance may have several transition points from one type of ordering to another.

As we are looking for possible antiferromagnetic arrangements of the A and B lattices separately, we have to subdivide the lattices further. The subdivision actually occurring is determined by the interactions responsible for the ordering. We assume that in both Aand B lattices the order is determined by the nearestneighbor interactions. The A lattice is then subdivided into two (A', A'') face-centered cubic lattices and the B lattice into four  $(B_1, B_2, B_3, B_4)$  face-centered cubic lattices, all having the same cube edge as the spinel cell. A site from A' has four sites from A'' as nearest neighbors in the A lattice. A site from  $B_1$  has six sites, two from each of the other three  $B_i$ 's, as nearest neighbors in the B lattice. The interactions between any two  $B_i$ 's are equal. In order to simplify the formalism we shall assume that only one type of magnetic ion is present. Actually, in a molecular field approximation, the more complicated case of several types of magnetic ions may be formally equivalent to our treatment.

<sup>&</sup>lt;sup>1</sup> L. Néel, Ann. phys. 3, 137 (1948).

<sup>&</sup>lt;sup>2</sup> For details of the crystal structure of ferrites, see E. J. W. Verwey and E. L. Heilmann, J. Chem. Phys. 15, 174 (1947).

## II. GROUND STATE AT 0°K

At absolute zero all sublattices are saturated. We show in the appendix that if we neglect anisotropy, the four sublattices  $B_i$  may be lumped together to form two equivalent sublattices, B' and B''. The molecular fields acting on lattices A', A'', B', B'' may be written:

$$\begin{aligned} \mathbf{H}_{a'} &= n(\alpha_1 \mathbf{M}_{a'} + \alpha_2 \mathbf{M}_{a''} - \mathbf{M}_{b'} - \mathbf{M}_{b''}), \\ \mathbf{H}_{a''} &= n(\alpha_2 \mathbf{M}_{a'} + \alpha_1 \mathbf{M}_{a''} - \mathbf{M}_{b'} - \mathbf{M}_{b''}), \\ \mathbf{H}_{b'} &= n(-\mathbf{M}_{a'} - \mathbf{M}_{a''} + \gamma_1 \mathbf{M}_{b'} + \gamma_2 \mathbf{M}_{b''}), \\ \mathbf{H}_{b''} &= n(-\mathbf{M}_{a'} - \mathbf{M}_{a''} + \gamma_2 \mathbf{M}_{b'} + \gamma_1 \mathbf{M}_{b''}), \end{aligned}$$

where  $\alpha_1$ ,  $\alpha_2$ ,  $\gamma_1$ ,  $\gamma_2$  are negative (antiferromagnetic) and  $\alpha_1 + \alpha_2 = 2\alpha$ ;  $\gamma_1 + \gamma_2 = 2\beta$ . The interaction constants n,  $n\alpha$  and  $n\beta$  are defined for a mole of magnetic ions distributed on lattices A and B in the ratio  $N_a/N_b$  $=\lambda/\mu = y$ , where y may vary from 0 to 1. Any magnetization configuration may be characterized by two angles  $\phi$  and  $\psi$  as shown in Fig. 1. The energy is:

$$E = -\frac{1}{2} \sum \mathbf{H}_{i} \cdot \mathbf{M}_{i}$$

$$= -n [\alpha_{1} M_{a'}^{2} + \alpha_{2} \mathbf{M}_{a'} \cdot \mathbf{M}_{a''} + \gamma_{1} \mathbf{M}_{b'}^{2} + \gamma_{2} \mathbf{M}_{b'} \cdot \mathbf{M}_{b''}$$

$$- (\mathbf{M}_{a'} + \mathbf{M}_{a''}) \cdot (\mathbf{M}_{b'} + \mathbf{M}_{b''})]$$

$$= -n M_{b'}^{2} [(\alpha_{1} - \alpha_{2} \cos 2\varphi)y^{2} + (\gamma_{1} - \gamma_{2} \cos 2\psi)$$

$$+ 4y \sin \varphi \sin \psi].$$

The values of  $\phi$  and  $\psi$  that minimize this expression are:

1. If  $\alpha_2 \gamma_2 > 1$ ,  $\phi = \psi = 0$ ; the doubly antiferromagnetic configuration is the lowest for all y.

2. If  $\alpha_2 \gamma_2 < 1$ , the lowest state will depend on y; the values of  $\gamma_2$  and  $1/\alpha_2$  determine three regions:

- (a) For  $0 < y < |\gamma_2|$ , the solution is  $\phi = \frac{1}{2}\pi$ ,  $\sin\psi$  $= y/|\gamma_2|$ .
- (b) For  $|\gamma_2| < y < 1/|\alpha_2|$ , the solution is  $\phi = \psi = \frac{1}{2}\pi$ .
- (c) For  $1/|\alpha_2| < y$ , the solution is  $\psi = \frac{1}{2}\pi$ ,  $\sin\phi$  $=1/(|\alpha_2|y).$

In the triangular configuration  $(a)^3$  the criterion  $y < |\gamma_2|$  means that the molecular field of B" acting on B' is larger than the field of A' (or A'') acting on B'. The equilibrium angle  $\psi$  is then such that lattice B' sees a total field (apart from its own) equal to the molecular field due to B''.

The triangular cases are in fact generalizations of Néel's solutions<sup>4</sup> III and IV. In case (a), the magnetization of lattice B is here  $2M_{b'}\sin\psi = 2Ma'/|\gamma_2|$ ; Néel gives  $2Ma'/|\beta|$ . The difference is due to the implicit assumption  $\gamma_2 = \beta$  made by Néel as a necessary consequence of his neglect of the antiferromagnetic arrangements: the condition for the paramagnetic state of lattice B to have as low an energy as an anti-



FIG. 1. Equilibrium configuration of the four sublattices.

ferromagnetic state is, from the expression for the energy, that no subdivision be possible such that  $\gamma_2 \neq \gamma_1$ ; then  $\gamma_1 = \gamma_2 = \beta$  follows. Actually  $\gamma_2$  has a value between  $\beta$  and  $4\beta/3$ , resulting in a smaller value for the magnetization of lattice B. Néel's interpretation for his solutions III and IV seems to be lack of saturation of lattices B and A respectively, rather than a triangular arrangement. This belief is based on his considerations on the variation of the total moment near 0°K: his graphical determination<sup>5</sup> of the effective field on lattice A leads to  $\mathbf{H}_a \rightarrow 0$  as  $T \rightarrow 0^{\circ} \mathrm{K}$ , a clearly erroneous result, which, however, is the only one consistent with paramagnetic lack of saturation. The variation with temperature of the total moment that he deduces<sup>6</sup> for cases III and IV has a non-zero slope at the origin, violating the third law of thermodynamics. This is a consequence of a non-zero entropy at 0°K, also due to lack of saturation.

We may notice here that it is not necessary to have two crystallographically different sites in order to have a triangular arrangement. The symmetry of a simple lattice may give rise to a triangular arrangement, as occurs in the plane triangular array which may be decomposed into three interpenetrating triangular lattices. With only nearest-neighbor antiferromagnetic interactions, the ground state has the three saturation magnetizations at 120° to each other, and is nondegenerate. Thus, the Ising model fails for this lattice.7 The molecular field treatment gives a ratio  $\theta/T_c = 2$  for nearest neighbor interactions. Here  $\theta$  is the constant appearing in the Curie-Weiss law  $\chi = C/(T+\theta)$ ;  $T_c$  is the transition temperature.

#### **III. CURIE POINTS**

Let C denote the molal susceptibility. In the disordered state the magnetic moments are given by:

$$M_{a'} = \lambda C H_{a'}/2T; \quad M_{b'} = \mu C H_{b'}/2T.$$

The vanishing of the characteristic determinant of the following four homogeneous equations yields the pos-

<sup>&</sup>lt;sup>3</sup> A similar consideration applies to configuration (c).

<sup>&</sup>lt;sup>4</sup> L. Néel, reference 1, p. 149.

<sup>&</sup>lt;sup>5</sup> L. Néel, reference 1, p. 153.

<sup>&</sup>lt;sup>6</sup> L. Néel, reference 1, p. 154. <sup>7</sup> G. H. Wannier, Phys. Rev. **79**, 357 (1950).



FIG. 2. A possible succession of transitions to various configurations in the same substance.

sible Curie points:

$$\begin{aligned} &\alpha_{1}M_{a'} + \alpha_{2}M_{a''} - M_{b'} - M_{b''} = (2T/\lambda Cn)M_{a'} \\ &\alpha_{2}M_{a'} + \alpha_{1}M_{a''} - M_{b'} - M_{b''} = (2T/\lambda Cn)M_{a''} \\ &- M_{a'} - M_{a''} + \gamma_{1}M_{b'} + \gamma_{2}M_{b''} = (2T/\mu Cn)M_{b'} \\ &- M_{a'} - M_{a''} + \gamma_{2}M_{b'} + \gamma_{1}M_{b''} = (2T/\mu Cn)M_{b''}. \end{aligned}$$

The four solutions for  $T_c$  are:

$$T_{c1} = \frac{1}{2}Cn\mu(\gamma_1 - \gamma_2),$$
  

$$T_{c2} = \frac{1}{2}Cn\lambda(\alpha_1 - \alpha_2),$$
  

$$T_{c3} = \frac{1}{2}Cn\{\alpha\lambda + \beta\mu \pm [(\alpha\lambda - \beta\mu)^2 + 4\lambda\mu]^{\frac{1}{2}}\}.$$

The — sign in  $T_{c3}$  gives a negative value and must be rejected. Again, we see that if  $\gamma_1 = \gamma_2$  and  $\alpha_1 = \alpha_2$ , only  $T_{c3}$  is left; this is Néel's result.<sup>8</sup>  $T_{c1}$  and  $T_{c2}$  are the antiferromagnetic Curie points of lattices *B* and *A*, respectively. The type of ordering below these upper Curie points is ferrimagnetic for  $T_{c3}$  and antiferromagnetic in the corresponding lattice for  $T_{c1}$  or  $T_{c2}$ .

We need to determine the highest  $T_c$  among the four solutions; depending on  $\lambda/\mu$  and the molecular field coefficients, several cases will occur.

The transition temperatures  $T_{c1}$  and  $T_{c2}$  are always positive. The criterion for a positive  $T_{c3}$ , given by Néel, is  $\alpha\beta < 1$ . Let us introduce two numbers K and L, both between 0 and 1, such that  $\alpha_2 - \alpha_1 = 2\alpha K$  and  $\gamma_2 - \gamma_1$  $= \frac{2}{3}\beta L$ . From the expressions for the  $T_c$ 's, the condition for  $T_{c3}$  to be the largest root may be found as follows:

- (a) If  $T_{c3} > T_{c2}$ , then  $(\lambda/\mu) < \{ [1-(K+1)\alpha\beta]/[K(K+1)\alpha^2] \}$  and  $[1-(K+1)\alpha\beta] > 0;$
- (b) If  $T_{c3} > T_{c1}$ , then  $(\lambda/\mu) > \{L(L+3)\beta^2/3[3-(L+3)\alpha\beta]\}$  and  $[3-(L+3)\alpha\beta]>0$ .



FIG. 3. Arbitrary configuration of the magnetizations on four  $B_i$  sublattices.

<sup>8</sup> L. Néel, reference 1, p. 145.

A value for  $\lambda/\mu$  can satisfy these equations if:

$$\frac{L(L+3)\beta^2}{3[3-(L+3)\alpha\beta]} < \frac{1-(K+1)\alpha\beta}{K(K+1)\alpha^2}.$$
(1)

This inequality reduces to  $\alpha\beta < 3/(K+1)(L+3)$ ; further,  $\alpha_2 = (K+1)\alpha$  and  $\gamma_2 = (L+3)\beta/3$  so that Eq. (1) becomes  $\alpha_2\gamma_2 < 1$ . Conversely, if  $\alpha_2\gamma_2 > 1$ , either  $3-(L+3)\alpha\beta < 0$ , or if  $3-(L+3)\alpha\beta > 0$ , the inequality sign in Eq. (1) has to be reversed. In either case  $T_{c3}$  is not the largest root. Thus, if the doubly antiferromagnetic arrangement is the ground state at  $0^{\circ}$ K, the upper Curie point is at either  $T_{c1}$  or  $T_{c2}$ . But if a triangular or a ferrimagnetic arrangement is the ground state, then the upper Curie point is not necessarily at  $T_{c3}$ . The possible cases are deduced from Eq. (1), whose left and right sides we designate by l and r, respectively.

(a)  $\alpha_2 \gamma_2 < 1$  with  $1 - (K+1)\alpha\beta > 0$  and  $3 - (L+3)\alpha\beta > 0$ : then

$$\begin{array}{lll} T_{c3} > T_{c1} \text{ and } T_{c3} > T_{c2} & \text{for } l < (\lambda/\mu) < r, \\ T_{c1} < T_{c3} < T_{c2} & \text{for } r < (\lambda/\mu), \\ T_{c2} < T_{c3} < T_{c1} & \text{for } (\lambda/\mu) < l. \end{array}$$

(b)  $\alpha_2 \psi_2 < 1$  with  $1 - (K+1)\alpha\beta < 0$  and  $3 - (L+3)\alpha\beta < 0$ : then for all  $\lambda/\mu$ ,  $T_{c3} < T_{c1}$  and  $T_{c3} < T_{c2}$ . There are no other possible cases when  $\alpha_2 \gamma_2 < 1$ . If  $\alpha_2 \gamma_2 = 1$ , then l = r and at  $(\lambda/\mu) = r$ , we have  $T_{c1} = T_{c2} = T_{c3}$ .

We shall now follow the substance down to  $0^{\circ}$ K and investigate the possibilities for lower Curie points, assuming the interactions to be independent of temperature. We notice that there is no close connection between the inequalities in (a) and (b) and the inequalities defining the ground state at  $0^{\circ}$ K. We expect, therefore, several possible cases, of which we will consider the simplest case and the most complicated case. Actually, there are five cases having from one to four transition points.

When  $T_{c3}$  is the largest root and the ferrimagnetic arrangement is the ground state, then only one transition, at  $T_{c5}$ , occurs.

When  $T_{c2}$  is the largest root and the ground state is the triangular arrangement with  $\phi = \frac{1}{2}\pi$  and  $\sin\psi = y/|\gamma_2|$ , we have four transitions, indicated in Fig. 2. This case is perhaps realized in mixed Zn-Ni ferrite with x=0.2. The transition temperature  $T_{c4}$  is calculated as follows: the triangular case gives  $\sin\phi = (M_{b'}/|\alpha_2|M_{a'})$ , where  $M_{a'}$  and  $M_{b'}$  are now the values of the moments at a temperature  $T < T_{c4}$ . Eliminating  $M_{a'} \sin\phi$ from the expression for  $H_{b'}$ , we get

$$H_{b'} = 2nM_{b'}(\beta + 1/|\alpha_2|).$$

The value of  $T_{c4}$  is given by using this expression in the limiting form of the Brillouin function for the magnetization:

$$M_{b'} = \mu C H_{b'}/2T$$

$$T_{c4} = \mu Cn(\beta + 1/|\alpha_2|)$$

whence

On the other hand,  $T_{c5}$  and  $T_{c6}$  may not be given in closed form.  $T_{c5}$  occurs when  $(M_{a'}/M_{b'}) = (1/|\alpha_2|)$  and  $T_{c6}$  when  $(M_{a'}/M_{b'}) = |\gamma_2|$ . These conditions allow their numerical or graphical determinations.<sup>9</sup>

We have treated so far the case  $\alpha_2 \gamma_2 < 1$ . For the case  $\alpha_2\gamma_2 > 1$ , we saw that the upper Curie point is at either  $T_{c1}$  or  $T_{c2}$ . Suppose it is at  $T_{c1}$ ; then the B lattice becomes ordered first. At  $T_{c2}$ , the A lattice which still sees no field from B, also becomes ordered antiferromagnetically and no other transition occurs down to 0°K. This case is not as interesting as the preceding one.

### IV. EXPERIMENTAL POSSIBILITIES

The existence of the triangular ground state may be ascertained by neutron diffraction experiments. The interaction constants n,  $n\alpha$ , and  $n\beta$  have been determined<sup>10</sup> for mixed Zn-Ni ferrites, Fe<sub>2</sub>O<sub>3</sub>, xNiO, (1-x)XZnO. The ratio of saturation magnetizations on lattices A and B is: y=x/[(2-x)+0.4x]; the value of  $\gamma_2$  is in between  $\beta$  and  $4\beta/3$ . For x=0.3, we obtain  $y \approx 0.16$  and  $\gamma_2 = 0.14$  to 0.18. The triangular arrangement is expected to appear in this neighborhood. The angle of the two components on the B lattice will be  $\approx 90^{\circ}$  at  $\sin \psi = (y/|\gamma_2|) = 0.7$ , or x = 0.2 to 0.25.

In general, because of the smallness of  $\beta$  in most ferrites, the triangular case will occur for rather small values of the saturation magnetization ratio y, of the order of  $|\beta|$ . As the molecular field treatment of the A-B and A-A interactions becomes less reliable with decreasing concentrations of the magnetic ions on  $A_{i}$ the criterion  $y < |\gamma_2|$  deduced above may be only a rough estimate.

Neutron diffraction could also ascertain the existence of antiferromagnetism in pure Zn ferrite and in mixed Zn ferrites. Magnetic measurements<sup>11</sup> on mixed Zn-Co and Zn-Mn ferrite yield data which, extrapolated to small concentrations of Co or Mn, suggest that  $T_{c3} \rightarrow 0$ for  $x \sim 0.2$ . For smaller x our theory predicts a doubly antiferromagnetic ground state. In this case too, the concentration of magnetic ions in lattice A is small.

The existence of the multiple Curie points should most easily be detected by heat capacity measurements. This method avoids the spurious effects in magnetic measurements caused by ferromagnetic impurities. We have estimated the discontinuities in heat capacity associated with the various types of transitions and find them to be well within the range detectable by current techniques. In antiferromagnetic transitions the contribution to the discontinuity (on the molecular field theory) due to the spin, is  $2.38 \ k$  per ion. In the triangular transition occurring at  $T_{c4}$  (see Fig. 2), the ions of lattice B contribute this same amount to the discontinuity, while the ions on lattice A do not contribute because the functional dependence of  $M_{a'}$  on



FIG. 4. Symmetrical configuration with the same energy.

 $H_{a'}$  does not change at the transition point. On the other hand, at transformations of the types associated with  $T_{c3}$ ,  $T_{c5}$  and  $T_{c6}$ , the functional dependences on  $H_{a'}$  and  $H_{b'}$  of both  $M_{a'}$  and  $M_{b'}$  change at the corresponding transformation points. The resulting discontinuities depend on the interaction parameters as well as on  $\lambda/\mu$ ; the expressions are rather complicated. As mentioned in Section III in connection with Fig. 2, mixed Zn-Ni ferrite with x < 0.3 is a promising substance for the investigation of the multiple transitions.

This research has been assisted in part by the Office of Naval Research. One of us (Y. Y.) is a Whiting Fellow of the University of California. Mr. Harvey Kaplan has kindly checked some of the calculations.

### APPENDIX

We show here that the energy of the lowest energy arrangement is the same with the B lattice subdivided into two sublattices as it is when B is subdivided into four sublattices. The effect of lattice A is equivalent to a field **H**. By symmetry,  $\sum \mathbf{M}_{bi}$  will be parallel to the direction of **H**. Consider the general case of Fig. 3: we will show that all configurations with the same resultant moment have the same self-energy, and also the same energy in the field **H**.

The self-energy is:

$$W = -\cos^2\varphi + \sin^2\varphi - \cos^2\psi + \sin^2\psi + 4\sin\varphi\sin\psi\cos(\alpha+\beta),$$

where the molecular field constant  $n\beta$  has been taken equal to -1. We consider now the reference state shown in Fig. 4, where  $\sin\varphi_0 = \sin\varphi \cos\alpha$  and  $\sin\psi_0$  $=\sin\psi\cos\beta$ . We have:

$$W_{0} = -\cos^{2}\varphi_{0} + \sin^{2}\varphi_{0} - \cos^{2}\psi_{0} + \sin^{2}\psi_{0} + 4\sin\varphi_{0}\sin\psi_{0}$$

and

$$W - W_0 = 2 \sin^2 \varphi \sin^2 \alpha + 2 \sin^2 \psi \sin^2 \beta$$
$$-4 \sin \varphi \sin \psi \sin \alpha \sin \beta$$

 $=2(\sin\varphi\sin\alpha-\sin\psi\sin\beta)^2=0$ 

as  $\sum \mathbf{M}_{bi}$  is in the direction of **H**. Furthermore,

$$W_0 = -2 + 2(\sin\varphi_0 + \sin\psi_0)^2$$

is a function only of  $\sin \varphi_0 + \sin \psi_0$ ; thus,  $M_{b3}$  and  $M_{b4}$ , and also  $M_{b1}$  and  $M_{b2}$ , may be taken collinear in pairs.

This freedom in the orientations of the  $M_{bi}$ 's is a result of the equality of the interaction constants. If

<sup>&</sup>lt;sup>9</sup> Néel gives a graphical method in Appendix I to his paper.
<sup>10</sup> L. Néel and P. Brochet, Compt. rend. 229, 1133 (1949).
<sup>11</sup> C. Guillaud and H. Creveaux, Compt. rend. 230, 1458 (1950).

we assume that the interactions 1-2 and 3-4 are -k, the other four interactions remaining -1, then at constant  $\sin \varphi_0 + \sin \psi_0$ , the lowest states are:

(a) for k < 1,  $M_{b1}$  and  $M_{b2}$ ,  $M_{b3}$  and  $M_{b4}$ , respectively, collinear;

(b) for k>1,  $M_{b1}$  and  $M_{b3}$  (or  $M_{b4}$ ),  $M_{b2}$  and  $M_{b4}$ (or  $M_{b3}$ ), respectively, collinear.

Thus, if the interactions in the *B* lattice are such that with no applied field B is subdivided into two oppositely magnetized sublattices B' and B'', then under the influence of A there will be no tendency for B' and B''

PHYSICAL REVIEW

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## Angular Correlation between Protons and Gammas in the $Li^{6}(d, p_{\gamma})Li^{7}$ Reaction\*

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The angular correlation between protons and gammas in the reaction  $Li^{6}(d, p\gamma)Li^{7}$  has been experimentally determined for the purpose of obtaining information on the 480-kev excited state of Li7. A bombarding energy of 500 kev was used, so that the Be<sup>8\*</sup> intermediate nucleus would be formed in the 22.5-Mev excited state reported by Whaling and Bonner. No correlation was observed within the statistical accuracy of the experiment.

#### INTRODUCTION

T has long been considered that the 480-kev excited state of Li<sup>7</sup> is a  ${}^{2}P_{\frac{1}{2}}$  state.<sup>1</sup> At least one shell model indicates the reasonableness of this assumption.<sup>2</sup> However, experimental evidence, such as the high probability of transition to this state in the  $B^{10}(n,\alpha)$  Li<sup>7</sup> reaction, has led to other hypotheses.<sup>3</sup> We felt it worthwhile to measure the angular correlation between protons and 480-kev gammas in the  $\text{Li}^6(d,p\gamma)$  Li<sup>7</sup> reaction in order to shed further light on the question. Any correlation would indicate a state other than  ${}^{2}P_{\frac{1}{2}}$ .

Presumably an angular correlation would be most pronounced if a single level in the compound nucleus is involved. Bombarding energy was chosen in relation to target thickness so that the Be<sup>8</sup> compound nucleus was formed with the energy of the 22.5-Mev excited state reported by Whaling and Bonner.<sup>4</sup>

#### METHOD

A Li<sup>6</sup>SO<sub>4</sub> (95 percent Li<sup>6</sup>) target was bombarded with 500-kev deuterons. Both proton and gamma-counters were of the scintillation type, using a thin anthracene

\* Supported by ONR and AEC. This work was part of a thesis submitted to the Graduate School of Rice Institute in partial fulfillment of the requirements for the Ph.D., June, 1951. A report was presented at the Houston meeting of the American Physical Society, November, 1951.

Society, November, 1951.
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<sup>1</sup> D. R. Inglis, Phys. Rev. 50, 783 (1936); E. Feenberg and E. Wigner, Phys. Rev. 51, 95 (1937).
<sup>2</sup> M. G. Mayer, Phys. Rev. 78, 16 (1950).
<sup>3</sup> D. R. Inglis, Phys. Rev. 74, 1876 (1948); B. T. Feld, Phys. Rev. 75, 1618 (1949).
<sup>4</sup> W. Whaling and T. W. Bonner, Phys. Rev. 79, 258 (1950).

layer for the protons and a large NaI crystal for the gamma-rays. The proton counter was at 90° to the deuteron beam, and the gamma-ray counter was rotated in the plane perpendicular to the beam.

to break into 4 (or, in general, into  $2^n$ ) sublattices, no

matter what the interactions between these sublattices

may be. This shows that our hypothesis about the

nearest-neighbor interactions determining the ordering

The neglect of anisotropy is justified only as far as

energy is concerned. Because of the degeneracy of the

arrangements with the same total moment on the B

lattice, anisotropy may lead to a configuration where

the four sublattices are magnetized all in different

is not an essential one and does not affect our results.

NaI was used because it gives a strong photoelectric peak. By using biased trigger circuits to eliminate pulses below this peak, it was possible to eliminate most of the gamma-ray background from the accelerator while eliminating only a minor fraction of the 480-kev gamma-rays from the target. It was felt that in this manner a lower percentage of accidental coincidences (experimentally 5 to 12 percent of the true count) was achieved than would have been possible with an organic phosphor, which is faster but does not allow strong energy selection.

The biased triggered circuits were blocking oscillators. The blocking oscillator pulses fed into the coincidence circuits were the same height and shape for all sizes of triggering pulse. This made possible a direct measurement of the accidental rate by using independent sources of protons and gammas equivalent in intensity to the target. A direct measurement by this method would have been difficult or impossible if raw pulses from the amplifiers had been fed directly into the coincidence circuit, since the independent gammasource would have necessarily had to give the same pulse-height distribution as the target.

## EXPERIMENTAL EQUIPMENT

The target was made by evaporation of the Li<sup>6</sup> enriched lithium sulfate onto a thin polished silver disk. Its thickness was 370 micrograms/cm<sup>2</sup> or about