The commutator in (11) is then expressible in terms of the anticommutator

$$\begin{bmatrix} \bar{\psi}(x)\psi(x),\psi(x') \end{bmatrix} = \bar{\psi}(x) \{\psi(x),\psi(x')\} - \{\bar{\psi}(x),\psi(x')\} \{\psi(x),\psi(x')\} \{\psi(x),\psi(x),\psi(x')\} \{\psi(x),\psi(x),\psi(x')\} \{\psi(x),\psi(x),\psi(x')\} \{\psi(x),\psi(x),\psi(x')\} \{\psi(x),\psi(x),\psi(x)\} \{\psi(x),\psi(x),\psi(x)\} \{\psi(x),\psi(x),\psi(x)\} \{\psi(x),\psi(x)\} \{\psi(x),\psi(x)\} \{\psi(x),\psi(x)\} \{\psi(x),\psi(x)\} \{\psi(x),\psi(x),\psi(x)\} \{\psi(x),\psi(x)\} \{\psi(x),\psi(x)\} \{\psi(x),\psi(x)\} \{\psi(x),\psi(x),\psi(x)\} \{\psi(x),\psi(x)\} \{\psi(x),\psi(x)\} \{\psi(x),\psi(x)\} \{\psi(x),\psi(x)\} \{\psi(x),\psi(x),\psi(x)\} \{\psi(x),\psi(x)\} \{\psi(x),\psi(x)\} \{\psi(x),\psi(x)\} \{\psi(x),\psi(x),\psi(x)\} \{\psi(x),\psi(x)$$

This enables one to determine the anticommutator function.

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## Generalizations of the Weiss Molecular Field Theory of Antiferromagnetism

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A Weiss field calculation has been carried out for antiferromagnetism in more complicated structures than the usual calculation allows, and has been shown to give results more detailed and more consistent with experimental evidence on the magnetic properties of such structures than does the simpler theory.

## I. INTRODUCTION. THE TWO-SUBLATTICE THEORY

SIMPLE theoretical treatment of antiferromagnetism, the phenomenon of antiparallel ordering of spins in a lattice, has been given by Van Vleck<sup>1</sup> and Néel.<sup>2</sup> This theory is very similar to the Weiss molecular field treatment of ferromagnetism. As Van Vleck points out, it is to a certain extent equivalent in rigor to this treatment; that is, to the first approximation of the Heisenberg theory of ferromagnetism. However, it should be pointed out that the problems encountered in a rigorous quantum theory of antiferromagnetism are greater than those in the ferromagnetic case. For instance, the lowest energy state which one would instinctively write down in this case is not an eigenstate of the Hamiltonian, and indeed even the energy at absolute zero is not correctly given by a molecular field treatment of antiferromagnetism, while the theory of ferromagnetism does not have this difficulty. For further discussion of these problems, the reader is referred to the work of Hulthén;3 we shall assume here that the molecular field theory is at least adequate in a semiquantitative way, as we shall indeed find it to be.

Van Vleck's theory is quite successful qualitatively; it explains why, above the "Curie temperature,"  $T_c$ , at which the spins lock in antiparallel, the susceptibility should follow a modified Curie law

$$\chi = C/(T+\theta)$$

with C the usual "free-ion" Curie constant. Below  $T_c$ the susceptibility should be slightly field-dependent, but should, for a powder sample, drop to a value twothirds of the  $T_c$  value by T=0. The predictions as to C and the susceptibility below the Curie point are roughly correct, but still a third prediction, that  $\theta = T_c$ , is very poorly satisfied in general. Néel modified the Van Vleck theory by introducing next nearest neighbor spin coupling, which as we shall show can explain some anomalous  $\theta/T_c$  ratios. However, the main object of the present paper is to show that Néel's procedure is too simple, and that, first, simply from internal consistency Néel cannot explain  $\theta/T_c$  ratios as large as some of those observed; second, that the simple theory is not applicable to most lattice structures encountered in actual antiferromagnets; and third, that the correct theory, taking into account lattice structure, is more consistent with the experimental results, particularly on  $\theta/T_c$ . The only difference in principle between the theory here presented and the earlier theories is in the matter of taking into account geometrical lattice structures; the basic approach remains that of the molecular field theory of Weiss.

I should like to thank Professor Schrödinger for

many valuable discussions in connection with this paper. Note added in proof:-The squared Klein-Gordon equation was proposed as a meson equation in the meantime by Bhabha (Phys. Rev. 77, 665 (1950)). For a discussion of the difficulties

of field theories with higher order equations see A. Pais and G. E.

Uhlenbeck, Phys. Rev. 79, 145 (1950).

Van Vleck's theory applies to lattices in which the magnetic ions can be divided into two sublattices, each sublattice having the property that it contains no nearest neighbors of its own members, but all of the nearest neighbors of the members of the other sublattice. Thus if we assume only nearest neighbor interactions, and use an effective field treatment, the effective field on one sublattice is given entirely by the average magnetization of the other sublattice. A typical example of a lattice which can be divided in this manner is the body-centered cubic lattice, in which one can assign the cube corners to one sublattice, the cube centers to the other.

The basic formula of the Weiss field approach is

$$\bar{S} = SB_s(\text{const.} \times H_{\text{eff}}/kT),$$
 (1)

<sup>&</sup>lt;sup>1</sup> J. H. Van Vleck, J. Chem. Phys. 9, 85 (1941). Van Vleck refers to previous papers by Bitter and Néel. <sup>2</sup> L. Néel, Ann. de Physique 3, 137 (1948). <sup>3</sup> L. Hulthén, Arkiv. f. Mat., Astr. o. Fys. 26A, No. 11 (1938).

(4)

or

where  $B_s$  is the Brillouin saturation function for a spin value S. For a simple exposition of the theory we shall need only the expansion of  $B_s$  for small values of H:

$$M = \text{const.} \times SB_{s}(\text{const.} \times H^{\text{eff}}/kT) \cong \gamma H^{\text{eff}}/T, \quad (2)$$

where all the constants are summarized in the symbol  $\gamma$ .

Let us write down the expression for the effective field, under the assumption that the effect of exchange is given by

$$V_{ij} = J \mathbf{S}_i \cdot \mathbf{S}_j, \tag{3}$$

 $V_{ij}$  being the interaction energy between spins *i* and *j*, *J* an exchange integral, and  $\mathbf{S}_i$  and  $\mathbf{S}_j$  the spins of the *i*th and *j*th atoms, which are neighbors. The Weiss field method replaces  $\mathbf{S}_i$  by  $\langle \mathbf{S}_i \rangle_{Av}$ , which is proportional to  $M_i$ ; the interaction then acts as an effective field due to the magnetization of the sublattice containing *i*. Thus we have two effective fields, one acting on each sublattice.

or

$$H_i^{\text{eff}} = H_0 - \lambda M_j \quad j \neq i.$$

 $H_1^{\rm eff} = H_0 - \lambda M_2, \quad H_2^{\rm eff} = H_0 - \lambda M_1,$ 

The quantity  $\lambda$  is proportional to the exchange integral J. We may write

$$\mathbf{M}_i = (\mathbf{H}_0 - \lambda \mathbf{M}_j) \boldsymbol{\gamma} / T.$$
 (5)

For the total magnetization we obtain

$$\sum_{i,2} \mathbf{M}_{i} = (2\mathbf{H}_{0} - \lambda \sum_{i,2} \mathbf{M}_{i}) \boldsymbol{\gamma} / T$$
(6)

and for the susceptibility

$$\chi = \frac{M_{\text{tot}}}{H_0} = \frac{(2\gamma/T)}{1 + (\lambda\gamma/T)} = \frac{2\gamma}{T + \theta} \bigg\}.$$

$$\theta = \lambda\gamma$$
(7)

The Curie point can be obtained easily by leaving out  $\mathbf{H}_0$  in Eq. (5). The resulting set of homogeneous equations will have a non-zero solution for  $\mathbf{M}$  at only one temperature, given by setting the determinant equal to zero.

$$\mathbf{M}_{i} + (\lambda \gamma / T_{c}) \mathbf{M}_{j} = 0,$$

$$\begin{vmatrix} 1 & \lambda \gamma / T_{c} \\ \lambda \gamma / T_{c} & 1 \end{vmatrix} = 0,$$
(8)

FIG. 1. Division of f.c.c. lattice into four sublattices.

$$T_c = \lambda \gamma = \theta. \tag{9}$$

An interesting point which can be made here is that  $T_c$  is always proportional to the maximum exchange energy one can obtain by assuming constant magnetization on each of our chosen sublattices. This is true since, in minimizing

(where  $H^{\text{eff}}$  now can be thought of as a linear operator acting on the quantity **M**), we solve the eigenvalue problem for the operator  $H^{\text{eff}}$ , which is essentially the same as solving the secular Eq. (8). The eigenvector **M** then gives the best energy arrangement, as well as that holding at  $T_c$ . This theorem enables us to check our choice of sublattices in more complicated cases, by trying to find arrangements which will give lower energies than the chosen set can afford.

## II. THE FACE-CENTERED CUBIC LATTICE

Many known antiferromagnetics crystallize in structures for which the preceding theory does not hold. The most common of these is the face-centered cubic lattice, which can be divided into no less than four simple cubic sublattices which have the property that the sublattice does not contain nearest neighbors. Figure 1 shows a sublattice division in a face-centered cubic structure; numbers 1, 2, 3, and 4 refer to the various sublattices. It will be seen that each sublattice contains four nearest neighbors in each of the other three. We shall now give a complete treatment of this arrangement.

Refer to the sublattices by the index n, ranging from 1 to 4. The interaction energy is

$$V_{nm} = 2ZJ\mathbf{S}_n \cdot \langle \mathbf{S}_m \rangle_{\mathsf{Av}} \tag{11}$$

where J is an exchange integral, and Z is the neighbor number, 4 for the f.c.c. lattice. Thus the effective field is

$$\mathbf{H}_{n}^{\text{eff}} = \mathbf{H}_{0} - (2JZ/g\beta) \sum_{m \neq n} \langle \mathbf{S}_{m} \rangle_{\text{Av}}, \qquad (12)$$

and the magnetization is

$$\mathbf{M} = \frac{1}{4} N g \beta \sum \mathbf{S}_n \tag{13}$$

where N is the number of atoms, g the gyromagnetic ratio, and  $\beta$  the Bohr magneton. We have the basic equation of the Weiss theory, corresponding to (2):

$$\bar{\mathbf{S}}_n = SB_s(\mathbf{y}_n) \tag{14}$$

$$y_n = (Sg\beta/kT)H_n^{\text{eff}}.$$
 (15)

If we limit ourselves at first to the region above the Curie point, we have small effective fields in general, and we can expand the Brillouin function in (14):

$$SB_s(y) \simeq \frac{1}{3}(S+1)y. \tag{16}$$

TABLE I. Constants of antiferromagnetic compounds.

Compound	Crystal structure	Paramagnetic lattice structure	No. of sublat- tices	θ	T e	$\theta/T$ e	$\left(\frac{x_T=0}{x_T=T_c}\right)$		ε/λ (Ou Lower	r theory) Higher
MnF <sub>2</sub>	Rutile	Body-centered rectangular	2	113°	72°	1.57	0.76	0.22		
FeF <sub>2</sub>	Rutile	Body-centered rectangular	2	117°	79°	1.48	0.72	0.19		
MnŌ	NaCl	f.c.c.	4	610°	122°	5.0	$\sim^{\frac{2}{3}}$		0.5	0.5
FeO	NaCl	f.c.c.	4	570°	198°	2.9	0.8		-0.02	+1.07
MnS	NaCl	f.c.c.	4	528°	165°	3.2			0.12	0.91
MnSe*	NaCl	f.c.c.	4	$\sim 435^{\circ}$	$\sim \! 150^\circ$	$\sim 3$				$\sim 1$
FeCl <sub>2</sub>	$CdCl_2$	Hexagonal layer structure	3	48°	23.5°	2.0)?		0.33		
CoCl <sub>2</sub>	$CdCl_2$	Hexagonal layer structure	3	38.1°	24.9°	1.53)?		0.21		
$NiCl_2$	$CdCl_2$	Hexagonal layer structure	3	68.2°	49.6°	1.37)?		0.16		

\* The Curie point of MnSe is hard to locate because of a large thermal hysteresis in this crystal, perhaps due to a phase transition (three known crystal structures exist). However, the most reliable data (Shull, private communication) is that given here.

Thus

$$\mathbf{S}_{n} = \frac{1}{3}S(S+1)(g\beta/kT)[\mathbf{H}_{0} - (2JZ/g\beta)\sum_{m\neq n}\mathbf{S}_{m}].$$
(17)

Sum from 1 to 4 to get the magnetization:

$$\sum_{1}^{4} \mathbf{S}_{n} = \frac{1}{3} S(S+1) (g\beta/kT) [4\mathbf{H}_{0} - (6JZ/g\beta) \sum_{1}^{4} \mathbf{S}_{n}]$$

or,

$$\sum_{1}^{4} \mathbf{S}_{n} = \frac{\frac{1}{3}S(S+1)(4g\beta/kT)H_{0}}{1 + [2JZS(S+1)/kT]}$$

and

$$\chi_{T>T_c} = \frac{M}{H_0} = \frac{\frac{1}{3}S(S+1)Ng^2\beta^2/k}{T+S(S+1)2JZ/k}$$
(18)

To find the Curie point we use no external field in (17) and find the point at which a non-zero solution of the following equation system is possible:

$$\mathbf{S}_{n} + \frac{1}{3}S(S+1)(2JZ/3kT) \sum_{m \neq n} \mathbf{S}_{m} = 0.$$
 (20)

The determinant is

$$\begin{vmatrix} 1 & x & x & x \\ x & 1 & x & x \\ x & x & 1 & x \\ x & x & x & 1 \end{vmatrix} = 0,$$

where

$$x = 2S(S+1)JZ/3kT.$$

Then

$$(1-x)^{3}(1+3x) = 0$$
  

$$T_{c} = 2S(S+1)JZ/3k = \theta/3.$$
 (21)

We can now find the susceptibility at the Curie point, which is

$$\chi_{T_c} = N g^2 \beta^2 / 8JZ. \tag{22}$$

The behavior of an antiferromagnetic f.c.c. lattice below the Curie point is rather complicated. We can see, by setting  $T = T_c$  in (20), that the only restriction on the spins is

$$\sum_{n} \mathbf{S}_{n} = \mathbf{0}.$$
 (23)

[It can easily be shown directly that (23) does indeed give the lowest energy for a system of n equally coupled spins.] However, (23) allows, for four spins, an infinite number of possible arrangements of spins, and the actual arrangement will no doubt be determined by next nearest neighbor interactions, anisotropy energy, etc. Since, as we shall see,  $\chi$  depends on the actual arrangement, it is impossible to find a unique value of  $\chi$  for  $T < T_c$ .We can, however, take two simple possible arrangements to show what forms for  $\chi$  are to be expected.

The first arrangement we choose is that in which two spins are parallel, two antiparallel:

The spontaneous magnetization can be determined from (14) and (12). Since

$$\sum_{n\neq n} \mathbf{S}_m = -\mathbf{S}_n,$$

$$S_n^0 = SB_s[(2SJZ/kT)S_n^0].$$
 (24)



FIG. 2.  $\theta/T_c$  for f.c.c. lattice with two interactions.

On expansion of all quantities about their values with mutually perpendicular. no external field we have

$$\frac{\mathbf{H}_{n}^{\circ} = \mathbf{H}_{n}^{\circ} + \delta \mathbf{H}_{n}}{\mathbf{S}_{n} = \mathbf{S}_{n}^{\circ} + \delta \mathbf{S}_{n}} \bigg\} .$$

$$(25)$$

Then

$$\delta H_n = H_0 - (2JZ/g\beta) \sum_{m \neq n} \delta S_m.$$
 (26)

We must calculate two cases:

$$\mathbf{H}_{0} \mid\mid \mathbf{S}_{0}, \tag{I}$$

$$\mathbf{H}_{0} \perp \mathbf{S}_{0}. \tag{II}$$

Case I. Expand the *B* function in the Taylor's series:

$$S_n^0 + \delta S_n = S[B_s(Sg\beta H_n^0/kT) + \delta H_n(Sg\beta/kT)B_s']$$
$$\delta S_n = \delta H_n(S^2g\beta/kT)B_s'$$
$$= [H_0 - (2JZ/g\beta)\sum_{m \neq n} \delta S_m](S^2g\beta/kT)B_s'$$

add up these equations for n=1 to 4:

$$\Sigma \delta S = \frac{4H_0(S^2 g\beta/kT)B_{s'}}{1 + (6JZS^2/kT)B_{s'}}$$
$$\chi_{II} = \frac{(Ng^2 \beta^2 S^2/kT)B_{s'}}{1 + (6S^2 JZ/kT)B_{s'}}$$
(27)

$$(\chi_{II})_{T=0} = 0$$
, since  $B_s'(\infty) = 0$ . (28)

Case II. Here we simply rotate the spins to be parallel to the effective field which they see. Thus

$$\begin{bmatrix} H_0 - (2JZ/g\beta) & \sum_{m \neq n} \delta S_m \end{bmatrix}$$
$$\delta S_n = |S^0| \,\delta H_n / |H_n^0| = |S_n^0| - \frac{(2JZ/g\beta) \sum_{m \neq n} \delta S_m^0}{(2JZ/g\beta) \sum_{m \neq n} S_m^0}$$

since all  $\delta S_n$  are presumably equal, this leads to

$$4\delta S_n = g\beta H_0/2JZ$$
  

$$\chi_{\perp} = Ng^2\beta^2/8JZ \qquad (29)$$

so that  $\chi_{\perp}$  is constant, and equal to  $\chi_{T_c}$  at T=0. Since, in averaging over the three directions for an isotropic sample, we take  $\chi_{\perp}$  twice and  $\chi_{\parallel}^{*}$  once, we get

$$(\chi_{\text{tot}})_{T=0} = \frac{2}{3} \chi_{T_c}.$$
 (30)

For this particular case we get the same result as that given by Van Vleck's simple theory.

A second case is that in which two pairs of spins are

S4 |\_\_\_\_\_  $S_2$ 

In case  $\mathbf{H}_0$  is perpendicular to all four, we obviously get the same result as in the previous case:

$$\chi_{\perp} = N g^2 \beta^2 / 8 J Z = \chi_{T_c}.$$

In case  $\mathbf{H}_0$  is parallel to the plane of the spins, we obtain the following:

$$\delta S_1 = \delta S_3, \quad \delta S_2 = \delta S_4 \delta S_1 = |S_0| \delta H_n / H_n^0 = |S_0| [H_0 - (2JZ/g\beta)(\delta S_1 + 2\delta S_2)] / (2JZ/g\beta) |S_0|$$

 $2\delta S_1 + 2\delta S_2 = (g\beta/2JZ)H_0.$ 

or

Thus

This leads to

$$\chi_{II} = N g^2 \beta^2 / 8JZ = \chi_{\perp} = \chi_{T_c}. \tag{31}$$

$$\chi_{\text{tot}} = \chi_{T_c}.$$
 (32)

Thus there are cases, for the face-centered cubic lattice, for which Van Vleck's relation

$$\chi_{T=0} = \frac{2}{3} \chi_{T_c}$$

is not necessarily true.

The comparison of our theory with experiment is shown in Table I. The data on several salts, taken primarily from Bizette's thesis,4 are presented, together with the crystal structure, the structure of the lattice of paramagnetic ions, and the number of sublattices assuming nearest neighbor interactions. Compounds with the CdCl<sub>2</sub> structure are included; here on the simple theory one would assume 3 sublattices and  $\theta/T_c = 2$ , but the data are not clear experimentally, and in addition it is not permissible to apply the Weiss field theory to essentially plane structures.

#### **III. INTRODUCTION OF NEXT NEAREST** NEIGHBOR INTERACTION

Néel<sup>2</sup> has explained the large  $\theta/T_c$  ratios sometimes observed by introduction of a next nearest neighbor interaction  $\epsilon$  as well as a nearest neighbor interaction  $\lambda$ into the simple two-lattice theory.  $\theta/T_c$  then becomes  $(1+\epsilon/\lambda)/(1-\epsilon/\lambda)$ . The  $\epsilon/\lambda$  ratios thus computed are given in Table I. We will show later how our theory can be similarly expanded; in most cases quite small  $\epsilon/\lambda$ values will suffice.

Some recent results obtained by Shull<sup>5</sup> at Oak Ridge

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<sup>&</sup>lt;sup>4</sup> H. Bizette, thesis (Massonand Company, Paris, 1946). See esp. p. 94. <sup>5</sup> Private communication.

with neutron diffraction seem at first to contradict the above theory. His measurements are made on MnO. First, a favorable point: he finds, at room temperature  $(<\theta)$ , a pattern corresponding to short-range order, which is physically very reasonable when one examines the mechanism by which  $\theta$  can become larger than  $T_c$ . Namely, the susceptibility (depending on  $\theta$ ) as well as the short-range order should, in first approximation, depend only on the number of neighbors and strength of coupling, whereas  $T_c$  is a lattice dependent quantity which measures the lowest energy obtainable with a given lattice. However, the present simple theory cannot give results on short-range order.

The observations of Dr. Shull at low temperatures  $(\langle T_c \rangle)$  do not confirm our ideas, however. He finds that an unexpected structure is present in which each of the four simple cubic sublattices is arranged anti-ferromagnetically within itself (as shown by the + and - signs on sublattice 1 in Fig. 1) while the sublattices are either completely uncorrelated, or are correlated in such a way that the spins in one set of (111) planes are all parallel. The neutron diffraction results cannot choose between these structures, nor is it possible to distinguish between them on our theory, as we shall see. We see at once that such a structure does not satisfy our theory, and the primary advantage which our theory has of explaining high  $\theta/T_c$  ratios without large  $\epsilon/\lambda$  ratios has been lost.

Let us see, nonetheless, what our theory does give in case there is a next nearest neighbor interaction. In this case it is necessary to subdivide the lattice still further, dividing each of the 4 sublattices into  $a + and a - sub-lattice, as is done in Fig. 1 for sublattice 1. We shall use the simplified notation of the first part of this paper, and work entirely with <math>T \ge T_c$ . Then we can write

$$\mathbf{M}_{n}^{\pm} = (\gamma/T) H_{n}^{\pm \mathrm{eff}} \tag{33}$$

and it is easy to see that if we assign an interaction  $\lambda$  to each nearest neighbor contact and  $\epsilon$  to each next nearest neighbor contact we obtain

$$\mathbf{H}_{n^{\pm \mathrm{eff}}} = \mathbf{H}_{0} - 2\lambda \sum_{m \neq n} (\mathbf{M}_{m^{\pm}} + \mathbf{M}_{m^{\mp}}) - 6\epsilon \mathbf{M}_{n^{\mp}}.$$
 (34)

We get for the various magnetizations

$$\mathbf{M}_{n}^{\pm} = (\boldsymbol{\gamma}/T) [\mathbf{H}_{0} - 2\lambda \sum_{m \neq n} (\mathbf{M}_{m}^{\pm} + \mathbf{M}_{m}^{\mp}) - 6\epsilon \mathbf{M}_{n}^{\mp}]. \quad (35)$$

To obtain the magnetization we sum over the 8 sublattices:

$$\sum_{n,\pm} \mathbf{M}_n^{\pm} = (\gamma/T) [8\mathbf{H}_0 - 12\lambda \sum \mathbf{M}_n^{\pm} - 6\epsilon \sum \mathbf{M}_n^{\pm}] \quad (36)$$

or,

$$\chi = 8\gamma H_0 / [T + 6\gamma(2\lambda + \epsilon)]$$
(37)

To obtain  $T_c$  we must solve the secular equation of the

following equation system:

$$\mathbf{M}_{n}^{\pm} + (2\gamma\lambda/T) \sum_{m \neq n} (\mathbf{M}_{m}^{\pm} + \mathbf{M}_{m}^{\mp}) + (6\gamma\epsilon/T) \mathbf{M}_{n}^{\pm} = 0. \quad (39)$$

The secular equation factors as follows:

$$= \left(1 - \frac{6\gamma\epsilon}{T}\right)^4 \left(1 + \frac{6\gamma\epsilon}{T} - \frac{4\gamma\lambda}{T}\right)^3 \left(1 + \frac{6\gamma\epsilon}{T} + \frac{12\gamma\lambda}{T}\right).$$

We have two choices for  $T_c$ :

$$\left. \begin{array}{c} T_{c1} = 6\gamma\epsilon, \\ T_{c2} = 4\gamma\lambda - 6\gamma\epsilon. \end{array} \right\}.$$

$$(40)$$

Then

0

$$\theta/T_{c1} = 1 + 2(\lambda/\epsilon), \\ \theta/T_{c2} = 3(1 + \epsilon/2\lambda)/[1 - 3\epsilon/2\lambda].$$

$$(41)$$

These two values of  $\theta/T_c$  are plotted as functions of  $\epsilon/\lambda$  in Fig. 2 as curve (1). Since we must, of course, always pick the highest value of  $T_c$ , or the lowest value of  $\theta/T_c$ , as the correct one, the solution changes at the point  $\epsilon/\lambda = \frac{1}{3}$ . We can solve Eq. (36) for the arrangement of *M*'s at the Curie point, and obtain

$$T_{c1}: \mathbf{M}_{m}^{+} + \mathbf{M}_{m}^{-} = 0$$
; no other restriction (42)

$$\left.\begin{array}{c}T_{c2}\colon \mathbf{M}_{m}^{+}-\mathbf{M}_{m}^{-}=0,\\\\\sum_{1}^{4}\mathbf{M}_{m}=0.\end{array}\right\}.$$
(43)

We see that  $T_{c1}$  corresponds to Shull's arrangement, which is perfect as far as next nearest neighbors go, but poor for nearest neighbors, while  $T_{c2}$  accommodates nearest neighbors but not next nearest neighbors. Thus in the region on the left of Fig. 2, nearest neighbor interaction predominates, while to the right next nearest neighbors are most important.

The case of MnO is not quite so easily accommodated as curve (1) indicates. It is possible to find lower energy configurations than that given by (43). We can see this by observing that if we set (for instance)  $S_1+S_2=0$  (so that  $S_3+S_4=0$ ) the nearest neighbor interaction between 100 planes as a whole vanishes. Thus we may pick a (100) direction, if we like, and change the direction of the spins on each plane at random going along this direction. The next nearest neighbor interaction will actually make it most favorable to change the sign of alternate planes in each sublattice so that the best sublattice structure when nearest neighbors are in control is not that of Fig. 1 but one in which sublattices 1 and 2, 3 and 4 exchange identities every other plane. The energy available now is proportional to  $(4\lambda - 2\epsilon)$ , rather than to  $(4\lambda - 6\epsilon)$  so that, by (44)

our theorem on the relation between  $T_c$  and the available energy, we know that the real  $T_c$  for the nearest neighbor region is

 $\theta/T_{c3} = 3(1+\epsilon/2\lambda)/[1-\epsilon/2\lambda]$ 

$$T_{c3} = \lambda (4\lambda - 2\epsilon), \qquad |$$

This is plotted as the dashed curve (2) of Fig. 2.

It will be seen that  $\theta/T_c = 5$  is still barely allowed by our theory, but that the  $\epsilon/\lambda$  ratio is definitely close to  $\frac{1}{2}$ for MnO. The two possible  $\epsilon/\lambda$  ratios for all compounds, under our theory, are included in Table I.<sup>6</sup> It is a nice confirmation of our theory that (a) MnO requires a high next nearest neighbor interaction simply because of its  $\theta/T_c$  value, while Shull's data confirm that next nearest neighbors are furnishing the antiferromagnetic alignment; (b) the theory of superexchange predicts that the next nearest neighbor interaction should increase along the series MnO-MnS-MnSe, as is ob-

<sup>6</sup> It was of interest to carry out the logical extension of the simple Néel two-sublattice theory for high values of the next nearest neighbor interaction: i.e., to divide each sublattice into two sublattices. The result is very like Fig. 2 except that  $\theta/T_c>3$ is not allowed in this case. For  $\epsilon > \lambda/2$ , the sublattices become antiferromagnetic, as in the f.c.c. structure. Thus simply on internal evidence alone the Néel theory cannot explain the  $\theta/T_c$ ratio in MnO.

served.<sup>7</sup> Shull has shown that MnSe has the "next nearest neighbor" arrangement of MnO, and thus the value  $\epsilon/\lambda = 1$  is to be preferred.

The reason for the large values of  $\epsilon/\lambda$  occurring in the theory must be sought for in a high superexchange<sup>7,8</sup> combined with a large separation of the magnetic ions in the antiferromagnetic crystals, leading to low direct exchange integrals. An examination of Kramers' theory indicates that if the superexchange is due to transitions of *p*-electrons from the negative to the positive ion (that is, to a partial covalent character of the bonds) the directionality of the superexchange must be that of a p-wave function; i.e., directly through the negative ion to the next nearest neighbor rather than the nearest neighbor. Also, since the other compounds should be expected to be less ionic than MnO, it is probable that their  $\epsilon/\lambda$  values are the higher ones,  $\cong$ 1, rather than the lower ones, as would be more satisfying from a naive picture.

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# $\mu$ -Pair Theories and the $\pi$ -Meson

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The  $\mu$ -pair theory suggests the interpretation of the  $\pi$ -meson as a pair of  $\mu$ 's bound together by a nucleon pair field. Only a non-relativistic description, involving a cut-off in the momentum spaces, is attempted. Various  $\pi$ -meson types, such as are well known from the conventional Yukawa theories, can be constructed, depending on the type of coupling adopted in the interaction Hamiltonian of the pair theory. It appears, however, that only the pseudoscalar coupling, which leads to a pseudoscalar  $\pi$ -meson, is consistent with the experimental data; e.g., on the nuclear scattering of  $\mu$ 's, which indicate that the interaction of  $\mu$ 's with nucleons, at least in such processes, is rather weak. Then, also, the creation of  $\mu$ -pairs in high energy nuclear collisions is expected to be an infrequent event, compared to the  $\pi$ -creation. Nonetheless, the  $\mu$ -pair production should furnish a crucial experimental test. Another process predicted is the dissociation of a fast  $\pi$ -meson, passing through matter, into a pair. The existence of heavier mesons, involving more than two  $\mu$ 's, seems likely.

## I. INTRODUCTION

 $\mathbf{A}^{\mathrm{S}}$  an alternative to Yukawa's theory, the pair theory of nuclear interactions was much discussed some years ago.1 The great advantage of this kind of theory is that it accounts for the saturation character of the nuclear forces without ad hoc assumptions.<sup>2</sup> The present paper is concerned with another

implication of the theory. According to  $\mu$ -pair theories,  $\mu$ -mesons may be bound together to form composite particles and, in particular,  $\pi$ -mesons may be interpreted as  $\mu$ -pairs. Thus, a unified picture of both meson specimens seems possible. Heavier mesons may also be foreseen.

It is true that the  $\mu$ -pair theory has been somewhat discredited lately by the lack of experimental evidence for a strong nuclear interaction of  $\mu$ -mesons. Even if only pairs of  $\mu$ 's are supposed to interact with nucleons, one expects at least a strong nuclear scattering of

<sup>&</sup>lt;sup>7</sup> P. W. Anderson, Phys. Rev. 79, 350 (1950). <sup>8</sup> H. A. Kramers, Physica 1, 182 (1934).

<sup>&</sup>lt;sup>1</sup> For the literature up to 1944, see W. Pauli and N. Hu, Rev.

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