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Spin Waves

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I. INTRODUCTION

THE problem of calculating the magnetization in a ferromagnetic, antiferromagnetic, or ferrimagnetic material is so complicated that it is always necessary to adopt some kind of approximation. Above the Curie temperature the partition function can be developed as a power series¹ in the ratio of the exchange integral to kT , but the convergence is exceedingly slow and each succeeding term is progressively more difficult to compute. A semiempirical, semitheoretical approach is provided by the Weiss molecular field method, or ramifications thereof, and this procedure works qualitatively even below the

Curie point. The Weiss-Bethe-Peierls method,¹ wherein the interaction inside a cluster is handled accurately but the coupling with its entourage phenomenologically, can be regarded as a refinement of the Weiss molecular field method.

At low temperatures, where the magnetization differs only slightly from that at absolute zero, none of these procedures is satisfactory. Instead, for a crystal composed of regularly spaced atoms, one has the method of spin waves, which is the subject of the present paper. Our purpose is twofold: firstly, to assemble in one place and in a unified fashion many of the results which are rather scattered in the literature, and secondly to introduce a simplified version of the quantum-mechanical theory. Our approach is based on approximating the magnetic spin system by a system of harmonic oscillators. The distinction from the conventional approach of Bloch,² and of Holstein and Primakoff,³

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¹ For references and a compendium of the most refined numerical results to date with the series and W-B-P methods see H. A. Brown and J. M. Luttinger, *Phys. Rev.* **100**, 685 (1955). In this connection reference should also be made to the easier, constant coupling approximation of P. W. Kasteleijn and J. Van Kranendonk, *Physica* **22**, 317 and 367 (1956).

² F. Bloch, *Z. Physik* **74**, 295 (1932).

³ T. Holstein and H. Primakoff, *Phys. Rev.* **58**, 1098 (1940).

based on creation and annihilation operators, is to a considerable extent only a semantic one, but nevertheless is probably of use to those readers to whom harmonic oscillators are more intuitive than the techniques of quantum-mechanical field theory.

Our treatment is based, except as otherwise stated, on the Heisenberg,⁴ or "localized-spin" model for a magnetic solid, which is analogous to the Heitler-London model of chemical bonds. The magnetism is regarded as coming entirely from electronic spins regularly spaced in the crystal. The question of the extent to which the model can be generalized to include itinerant effects of the magnetic electrons, i.e., conduction band structures, or to allow for the fact that actually the number of spins per lattice site is not integral in most ferromagnetic materials, is examined briefly in Sec. XI. In our opinion the extension to such cases is either quite involved or of only qualitative validity, so that in a certain sense the conventional spin-wave theory is only academic because it is based on a too idealized model. This objection does not, however, apply in equal degree to the application to nonconducting antiferromagnetics or ferrites.

It is perhaps well to take stock at the outset as to what results are obtained from the spin-wave theory, if questions of the applicability of the model are waived. In ferromagnetics, formulas are obtained for the approach of the magnetization to the value at absolute zero (perhaps the most celebrated result), for the dependence of the magnetization on field strength, for an exchange specific heat at low temperatures, and for the frequency in ferromagnetic resonance. The inclusion of the terms neglected in the usual spin-wave treatment, but still based on the idealized Heisenberg or Heitler-London model, furnish a mechanism for the existence of a spin-spin relaxation time. Corresponding results are also supplied by the spin-wave theory for antiferromagnetics, though here the approximations are more open to question on scores of rigor.

The literature of spin-wave theory is considerable. About fifty references are included in the present article, and even so our bibliography is probably not complete. Two different methods have been used to introduce the concept of spin waves, a quantum-mechanical and a semiclassical approach. The quantum-mechanical method was initiated by Slater⁵ and by Bloch² in attempts to derive approximate expressions for the low-lying energy levels of a ferromagnetic crystal. A different procedure, leading to essentially the same results, was later introduced by Holstein and Primakoff.³ The semiclassical method was developed by Heller and Kramers⁶ with the aim of giving a classical interpretation of the spin waves

introduced by Slater and Bloch. Both methods have recently been reviewed by Keffer, Kaplan, and Yafet⁷ in connection with ferro and antiferromagnetic resonance.

We now describe in mathematical terms the localized spin model which we use. If we include only isotropic spin coupling, as in the original Bloch theory, the Hamiltonian function of the spin system is

$$\mathcal{H} = -Hg\beta \sum_i S_{iz} - 2J \sum_{\text{nei}} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (1)$$

where β is the Bohr magneton $e\hbar/2mc$, g is the spectroscopic splitting factor, approximately equal to 2, and \mathbf{S}_i is the spin vector of atom i , measured in multiples of \hbar . It is assumed that all magnetic atoms have the same spin quantum number S . The first term of (1) is the Zeeman energy of the spins in a field of magnitude H , which throughout we suppose directed along the z axis. The second term is the exchange energy; the proof that it is proportional to the scalar product of the two spin vectors is quite standard⁸ and is omitted here. We restrict ourselves to simple lattices, and suppose the exchange coupling is negligible except between atoms which are nearest neighbors. The only nonvanishing exchange integrals then have a common value J , and the summation in (1) extends only over neighboring atoms, as indicated by the subscript nei , a convention used throughout the paper. The coupling constant J , which is the same as the familiar exchange integral between adjacent atoms, is positive for ferromagnetics, negative for antiferromagnetics. The more general case where the exchange energy takes the form $-2\sum_{j>i} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j$, and the sum runs over all pairs of atoms in the crystal, regardless of relative distance, can still be treated by spin waves if the lattice is regular and boundary effects are disregarded, but we shall not give calculations explicitly for this generalization; the procedures would be similar to those which we use.

All calculations with spin waves are based on the vector or Heisenberg model, wherein the coupling is proportional to the scalar product $\mathbf{S}_i \cdot \mathbf{S}_j$. Ising⁹ instead arbitrarily used a potential $S_{iz}S_{jz}$. From a mathematical standpoint this is a simplification, as then each individual S_{iz} is a constant of the motion and can be quantized. However, the exchange effects of quantum mechanics actually lead to the vectorial form. Hence the Ising model is a purely academic abstraction. It does not, in fact, give any spin waves at all, and the whole point of the spin-wave theory is essentially to handle a situation more real than that corresponding to the Ising model.

Besides the isotropic coupling included in (1)

⁷ Keffer, Kaplan, and Yafet, *Am. J. Phys.* **21**, 250 (1953).

⁸ See, for instance, J. H. Van Vleck *Electric and Magnetic Susceptibilities* (Oxford University Press, New York, 1932), Chap. XII, where the Dirac vector model and the standard Heisenberg theory are reviewed.

⁹ E. Ising, *Z. Physik* **31**, 253 (1925). For a review of literature on the Ising model see G. F. Newell and E. W. Montroll, *Revs. Modern Phys.* **25**, 353 (1953).

⁴ W. Heisenberg, *Z. Physik* **49**, 619 (1928).

⁵ J. C. Slater, *Phys. Rev.* **35**, 509 (1930).

⁶ G. Heller and H. A. Kramers, *Proc. Roy. Acad. Amsterdam* **37**, 378 (1934). See also N. J. Klein and R. S. Smith, *Phys. Rev.* **80**, 1111 (1950) and W. Döring, *Z. Physik* **124**, 501 (1947).

(so-called because the scalar product $\mathbf{S}_i \cdot \mathbf{S}_j$ depends only on the angle between the vectors \mathbf{S}_i and \mathbf{S}_j and is independent of the orientation of their resultant), there are actually anisotropic interactions. Those of the lowest order are of the "dipolar" or "tensor" structure

$$\sum_{j>i} D_{ij} [(\mathbf{S}_i \cdot \mathbf{S}_j) - 3(\mathbf{S}_i \cdot \mathbf{r}_{ij})(\mathbf{S}_j \cdot \mathbf{r}_{ij})/r_{ij}^2]. \quad (2)$$

For classical electromagnetic, i.e., true magnetic spin-spin interaction, the constant D_{ij} has the value

$$D_{ij} = g^2 \beta^2 / r_{ij}^3. \quad (3)$$

However, it is now generally recognized that nonclassical values of D_{ij} , and of much larger magnitude at short interatomic distances, can result from indirect repercussions of spin-orbit coupling, by a complicated perturbation mechanism¹⁰ which we do not describe here. Such nonclassical values of D_{ij} are often designated as "pseudodipolar" or "anisotropic exchange" coupling. The most important effects of the classical dipolar coupling are long-range. Hence when the value (3) of D_{ij} is used in (2), it is essential that the summation be unrestricted rather than confined to nearest neighbors. On the other hand, the nonclassical part of D_{ij} is of comparatively short range, like J_{ij} .

The expression (2) represents the most general anisotropic coupling between two atoms if their spins are each $\frac{1}{2}$, and if there is symmetry around the line of centers connecting them. For spins greater than $\frac{1}{2}$, there can also be quadrupolar coupling terms such as

$$E_{ij} (\mathbf{S}_i \cdot \mathbf{r}_{ij})^2 (\mathbf{S}_j \cdot \mathbf{r}_{ij})^2.$$

Still higher moments can also appear for greater spin values. The quadrupolar interaction can be important for ferromagnetic anisotropy,¹⁰ inasmuch as it can give rise to anisotropy in a cubic crystal even in first order. However, interaction of the dipolar structure (2), though capable, even in first approximation, of giving anisotropy in a noncubic crystal, can do so only in the second approximation in a cubic material. This property is an immediate consequence of symmetry requirements, since a quadratic form in the direction cosines, such as results from any first-order calculation based on (2), degenerates into a spherically symmetric affair when all three axes are equivalent. On the other hand, when squared, as in a second-order perturbation calculation, the biquadratic angular dependence $\lambda^2 \mu^2 + \lambda^2 \nu^2 + \mu^2 \nu^2$ characteristic of cubic anisotropy can be obtained.

We do not include quadrupolar coupling in any of our calculations, although it may be implicitly reflected in the artifice of an effective anisotropy field used in some of the later sections of the paper. For simplicity we defer until Sec. V the inclusion of the dipolar interaction (2), which is important for ferromagnetic resonance and for demagnetization effects.

II. SPIN-DEVIATION QUANTUM NUMBERS

Following Holstein and Primakoff³ we introduce, instead of the usual quantum number m which corresponds to the z component of the atomic spin \mathbf{S} , the spin-deviation quantum number $n = S - m$, which measures the deviation of S_z from its maximum value S . The allowed values of n are 0, 1, 2, \dots , $2S$, and these are the eigenvalues of the spin-deviation operator $n = S - S_z$. In a representation in which this operator n is diagonal, the nonvanishing matrix elements of the three components of \mathbf{S} are given by the familiar expressions

$$\begin{aligned} \langle n | S_x | n+1 \rangle &= \langle n+1 | S_x | n \rangle = \frac{1}{2} (n+1)^{\frac{1}{2}} (2S-n)^{\frac{1}{2}}; \\ \langle n | S_y | n+1 \rangle &= \langle n+1 | S_y | n \rangle^* \\ &= -\frac{1}{2} i (n+1)^{\frac{1}{2}} (2S-n)^{\frac{1}{2}}; \quad (4) \\ \langle n | S_z | n \rangle &= S - n. \end{aligned}$$

The matrices of the components of \mathbf{S}_i have the form (4) with respect to n_i , but are diagonal with respect to the n_j of all the other atoms in the crystal. Thus the matrices of \mathbf{S}_i are the direct products of the matrices (4) and $N-1$ unit matrices.

In the equilibrium state at absolute zero, all the spins of a ferromagnetic crystal are aligned parallel to the external field, so that no spin deviations are present and all the n_i vanish. It is well known that if in this state one introduces one spin deviation on a particular atom, this spin deviation does not remain localized on that atom: because of the exchange interaction with the surrounding atoms it will propagate itself through the lattice, thus constituting a "spin wave," or rather a spin-wave packet. A spin wave corresponds to the propagation of a spin deviation with a definite wave-number vector. The states of the spin system which correspond to excitation of spin waves of definite wave numbers are exact eigenstates of the Hamiltonian (1) as long as there is only one spin deviation present in the whole lattice, as we show later. In other words, if we use primes to denote the resultant spin of the entire crystal, which is, of course, the vector sum $\mathbf{S}' = \sum_i \mathbf{S}_i$ of the spins of the individual atoms, the eigenvalues of (1) can be computed exactly for $S'_z = NS$ and $S'_z = NS - 1$, where N is the total number of magnetic atoms in the crystal.

The essence of the whole spin-wave approximation is that for small values of the total crystalline spin deviation, the difference in energy between a state with $S'_z = NS - n$ and the completely parallel one $S'_z = NS$ can be regarded as approximately equal to the sum of n rigorously computed differences for unit deviations. In other words, the assumption of additivity is made in connection with spin reversals. Actually, this assumption is not accurate. If, for instance, two spin deviations are present, the energy of the spin system will depend on whether the spin deviations are located (i) on the same atom (this is possible only for $S > \frac{1}{2}$), (ii) on neighboring atoms, or (iii) on more

¹⁰ J. H. Van Vleck, Phys. Rev. 52, 1195 (1937).

distant atoms the spins of which are not coupled in the Hamiltonian (1). Because of this dependence of the total energy on the separation between spin deviations, spin waves interact with each other, and therefore do not satisfy the superposition principle accurately. There are two different causes for error. If we try to visualize the resulting corrections to the energy, one type of correction can be interpreted as a repulsion, the other as an attraction between spin deviations. For $S = \frac{1}{2}$, for instance, the repulsion results from the fact that there cannot be more than one spin deviation on a given atom. Thus if two spin deviations approach the same lattice site, they will be scattered because of this "interaction." If $S > \frac{1}{2}$, this repulsion appears only if there are more than $2S$ spin waves present, and the situation is then more complicated. The attractive part of the interaction arises because the total exchange energy of a configuration in which two spin deviations are located on nearest neighbors, is lower than that of a configuration in which the two spin deviations are farther apart. In a one-dimensional lattice, this attraction can give rise to "spin complexes" (cf. Bethe),¹¹ i.e., to states in which the spin deviations are bound to each other, and of course also to scattering. It should be clearly understood, however, that the total scattering cross section and the resulting correction to the energy are determined by the combined action of the repulsive and attractive interactions.

The basis of the *spin-wave approximation* is the fact that at sufficiently low temperatures the influence of the interaction between the spin deviations may be neglected. At low temperatures the magnetization deviates but slightly from the saturation magnetization. The average number of spin deviations in the crystal is then small compared to the total number of atoms in the crystal, and their interaction may then be neglected.

III. HARMONIC OSCILLATOR APPROXIMATION FOR FERROMAGNETISM

The similarity between the spin matrices (4) and the well-known matrices of the coordinate and momentum of a harmonic oscillator is the starting point of the present treatment of spin waves. In this section the method is explained for the ferromagnetic case without the dipolar correction (2). The antiferromagnetic case is discussed in Secs. VII-VIII.

Consider a linear harmonic oscillator of mass m and angular frequency ω . The nonvanishing matrix elements of the coordinate x and the momentum p of this oscillator are given by

$$\begin{aligned}\langle n | x | n+1 \rangle &= \langle n+1 | x | n \rangle = (\hbar/2m\omega)^{\frac{1}{2}}(n+1)^{\frac{1}{2}}, \\ \langle n | p | n+1 \rangle &= \langle n+1 | p | n \rangle^* = -i(\hbar m\omega/2)^{\frac{1}{2}}(n+1)^{\frac{1}{2}},\end{aligned}\quad (5)$$

where n now corresponds to the number of quanta

with which the oscillator is excited, i.e.,

$$\left\langle n \left| \frac{1}{2m} p^2 + \frac{1}{2} m \omega^2 x^2 - \frac{1}{2} \hbar \omega \right| n \right\rangle = n \cdot \hbar \omega. \quad (6)$$

When we introduce the dimensionless variables

$$Q = (m\omega/\hbar)^{\frac{1}{2}} x \quad \text{and} \quad P = (\hbar m\omega)^{-\frac{1}{2}} p, \quad (7)$$

we can write (5) and (6) in the following form:

$$\begin{aligned}\langle n | S^{\frac{1}{2}} Q | n+1 \rangle &= \langle n+1 | S^{\frac{1}{2}} Q | n \rangle \\ &= \frac{1}{2}(n+1)^{\frac{1}{2}}(2S)^{\frac{1}{2}}; \\ \langle n | S^{\frac{1}{2}} P | n+1 \rangle &= \langle n+1 | S^{\frac{1}{2}} P | n \rangle^* \\ &= -\frac{1}{2}i(n+1)^{\frac{1}{2}}(2S)^{\frac{1}{2}}; \\ \langle n | S - \frac{1}{2}(P^2 + Q^2 - 1) | n \rangle &= S - n.\end{aligned}\quad (8)$$

Comparing these expressions with the expressions (4) for the matrix elements of the spin components, we see that there is a striking similarity between the two sets of matrices. Both sets have nonvanishing matrix elements only on or immediately along the diagonal, and the values of these matrix elements are closely related. There are, however, two important differences, viz.:

(i) The matrices (8) are of infinite dimension, while the spin matrices (4) are only $(2S+1)$ -dimensional. An alternative way of expressing this is to say that the spin matrices (4) are, like (8), of infinite dimension, but all matrix elements corresponding to $n > 2S$ now being equal to zero rather than being equal to the values (8).

(ii) The factor $(2S-n)^{\frac{1}{2}}$ appearing in the spin matrices (4) is replaced by $(2S)^{\frac{1}{2}}$ in the harmonic oscillator matrices (8). We note that the matrix elements connecting the first two states $n=0$ and $n=1$ in (4) are equal to those in (8) for any value of S , while for larger values of n corresponding elements are approximately equal as long as n is small compared to $2S$.

Let us now replace in the Hamiltonian (1) of the spin system the spin matrices (4) by the harmonic oscillator matrices (8), i.e., let us carry out the following substitutions:

$$S_x = S^{\frac{1}{2}} Q; \quad S_y = S^{\frac{1}{2}} P; \quad S_z = S - \frac{1}{2}(P^2 + Q^2 - 1). \quad (9)$$

We then get the following Hamiltonian:

$$\begin{aligned}\mathcal{H} = & -Hg\beta \sum_i [S - \frac{1}{2}(P_i^2 + Q_i^2 - 1)] \\ & - 2JS \sum_{\text{nei}} (P_i P_j + Q_i Q_j) + \\ & - 2J \sum_{\text{nei}} [S - \frac{1}{2}(P_i^2 + Q_i^2 - 1)] \\ & \times [S - \frac{1}{2}(P_j^2 + Q_j^2 - 1)].\end{aligned}\quad (10)$$

The properties of the system of coupled harmonic oscillators described by the Hamiltonian (10) are the same as those of the spin system (1) as long as the temperature is so low that the states $n_i \geq 2$ of each of the oscillators are not appreciably excited. At higher temperatures, however, deviations appear. For $S = \frac{1}{2}$,

¹¹ H. A. Bethe, Z. Physik 71, 205 (1931).

neglect of the repulsive interaction is not justified when the levels with $n \geq 2$ become appreciably excited, while for larger values of S additional corrections occur because at higher temperatures the factor $(2S-n)^{\frac{1}{2}}$ may no longer be replaced by $(2S)^{\frac{1}{2}}$. These deviations of the properties of the oscillator system (8) from those of the spin system (1) are the result of the "anharmonicity" of the spin variables, i.e., to the deviations of the spin matrices (4) from the harmonic oscillator matrices (8).

One can improve the approximation (9) by adding higher-order (anharmonic) terms in P and Q , so as to make not only the matrix elements corresponding to $n=0$ and $n=1$ agree, but also those corresponding to $n=2$, etc., each additional term in (9) taking care of the next set of matrix elements corresponding to the next higher value of n . We shall illustrate this procedure for $S=\frac{1}{2}$. When we introduce the quantities $S^{\pm}=S_x \pm iS_y$, $a=2^{-\frac{1}{2}}(Q+iP)$ and $a^*=2^{-\frac{1}{2}}(Q-iP)$, we can write the first approximation (9) in the form

$$S^+=(2S)^{\frac{1}{2}}a; \quad S^-=(2S)^{\frac{1}{2}}a^*; \quad S_z=S-a^*a. \quad (11)$$

Only the first four matrix elements, those between the states $n=0$ and $n=1$, of the right-hand members of (11) are equal to the corresponding matrix elements of the left-hand members. In the next approximation, we can make the first nine matrix elements, corresponding to the states $n=0, 1$, and 2 , agree by replacing (11) by

$$S^+=(2S)^{\frac{1}{2}}(a-a^*aa); \quad S^-=(2S)^{\frac{1}{2}}(a^*-a^*a^*a); \quad (12)$$

$$S_z=S-a^*a+a^*a^*aa;$$

as can be verified easily by writing out the matrices occurring in (12). For $S > \frac{1}{2}$, the factor $(1-n/2S)^{\frac{1}{2}}$ appearing for $n \leq 2S$, and the repulsion now appearing for $n > 2S$, can be taken into account in a similar way. In the literature, a number of attempts have been reported¹²⁻¹⁴ to improve the approximation (11) by expanding the square root $(1-n/2S)^{\frac{1}{2}}$ in powers of $n/2S$ and retaining only the first few terms, but ignoring the "repulsive" interaction corresponding to the vanishing of the spin matrices (4) for $n > 2S$. In this way all the matrix elements corresponding to $n \leq 2S$ are partly corrected (this makes sense only for $S > \frac{1}{2}$), whereas in the present method (applicable also for $S=\frac{1}{2}$) the successive approximations (11), (12), etc., make successive sets of matrix elements have exactly the correct values (4). It should be cautioned, however, that it is not certain that this approximation procedure converges. We shall not discuss this point here, since we are at present interested only in the conventional spin-wave approximation.

The customary spin-wave approximation is obtained by neglecting not only the anharmonicity of the spin

variables, i.e., by using the first approximation (9), but by neglecting in addition the attractive interaction between the spin deviations. This interaction is represented by the fourth-order terms appearing in the last term of the Hamiltonian (10). The fact that the fourth degree part of (10) has a negative sign for $J > 0$ means that this neglected part of (10) can be interpreted physically as an attraction between the spin deviations. When these terms are neglected, the Hamiltonian (10) becomes

$$\mathcal{H}=E_0'+g\beta(H+H_E)\sum_i \frac{1}{2}(P_i^2+Q_i^2-1)+ \\ -2JS\sum_{\text{nei}}(P_iP_j+Q_iQ_j), \quad (13)$$

with

$$H_E=2JSz/g\beta.$$

Here, and throughout the article, z denotes the number of nearest neighbors of a given atom. H_E is the familiar molecular field at saturation, and the constant

$$E_0'=-NHg\beta S-\frac{1}{2}Nz \cdot 2JS^2, \quad (14)$$

is the energy of the completely saturated state in which all the spins are parallel to the external field.

As we see from (13), the Hamiltonian (1) is now reduced to the Hamiltonian of a system of N coupled harmonic oscillators described by the (reduced) coordinates and momenta Q_i and P_i . Except for the last term of (13), which represents a coupling between neighboring oscillators (carefully to be distinguished from the interaction between neighboring spin deviations, which we neglect), the expression (13) is exactly of the form one would expect on a naive basis, as we shall now show. Since we neglect the interaction between the spin deviations, we may suppose that each spin deviation is always surrounded by atoms without spin deviations. The difference in energy between a state in which there is one spin deviation on a particular atom and the corresponding state in which there is no spin deviation on that atom is then given by

$$\Delta E=[-Hg\beta(S-1)-2JzS(S-1)] \\ -[-Hg\beta S-2JzS^2]=g\beta(H+H_E), \quad (15)$$

if we neglect the influence of the x and y components of the spin, which are responsible for the coupling term in (13). When we want to replace the spin by a harmonic oscillator, we must require that the energy necessary to excite the oscillator to its first excited state be equal to the energy necessary to introduce one isolated spin deviation, i.e., we must put the $\hbar\omega$ of the oscillator equal to the energy (15). Neglecting again the coupling between neighboring oscillators, we would then arrive at the Hamiltonian

$$\mathcal{H}_0=\sum_i \left(\frac{1}{2m}p_i^2 + \frac{1}{2}m\omega^2x_i^2 - \frac{1}{2}\hbar\omega \right) + E_0', \quad (16)$$

where E_0' is given by (14) and is equal to the energy when all the oscillators are in their ground state.

¹² J. M. Ziman, Proc. Roy. Soc. (London) A65, 540 and 548 (1952).

¹³ R. Kubo, Phys. Rev. 87, 568 (1952).

¹⁴ M. R. Schafroth, Proc. Phys. Soc. (London) A67, 33 (1953).

When we scale the coordinates and momenta in accordance with (7), and put $\hbar\omega$ equal to the energy (15), the expression (16) becomes identical with (13), except that the coupling term, i.e., the term involving the sum over neighbors, is missing. This term is blithely disregarded in the Ising model,⁹ but is all important for the present article or any other treatment of spin waves.

The problem of finding the normal modes of the system of coupled harmonic oscillators is a simple one, the desired transformation being the well-known Slater transformation,⁵ either to the running waves

$$\begin{aligned} Q_k &= (1/N)^{1/2} \sum_i \exp(i\mathbf{k} \cdot \mathbf{r}_i) Q_i; \\ P_k &= (1/N)^{1/2} \sum_i \exp(-i\mathbf{k} \cdot \mathbf{r}_i) P_i; \end{aligned} \quad (17)$$

or to the real standing waves

$$k_z \geq 0 \begin{cases} Q_k = (2/N)^{1/2} \sum_i \cos(\mathbf{k} \cdot \mathbf{r}_i) Q_i; \\ P_k = (2/N)^{1/2} \sum_i \sin(\mathbf{k} \cdot \mathbf{r}_i) P_i; \end{cases} \quad (18a)$$

$$k_z < 0 \begin{cases} Q_k = (2/N)^{1/2} \sum_i \sin(\mathbf{k} \cdot \mathbf{r}_i) Q_i; \\ P_k = (2/N)^{1/2} \sum_i \cos(\mathbf{k} \cdot \mathbf{r}_i) P_i. \end{cases} \quad (18b)$$

The choice of the proper values of \mathbf{k} requires some comment. It is customary to introduce the so-called periodic or von Karman boundary conditions which imply that the wave function reverts to its original value after traversing the lattice. Expressed differently, the imposition of this type of boundary condition implies that the spins at the very edges of the crystal have neighbors rather than being without them on one side. This unreal supposition is made in order to place all spin situations on a par and so give full periodicity to the problem, with resulting simplification in the analysis. As long as the crystal is large,¹⁵ the error involved in using such boundary conditions is well known to be negligible, unless there are long range forces. (Dipolar interaction, however, can cause trouble as it vanishes only as $1/r^3$.) With the periodic boundary conditions, the allowed values of the vector $\mathbf{k}/2\pi$ are represented in general by points in the so-called reciprocal lattice for the crystal. For a simple cubic lattice, and a cubic crystal of edge L , the permitted values of k_x , k_y , and k_z are integral multiples of $2\pi/L$.

In terms of the standing waves amplitudes (18), the Hamiltonian (13) takes the form

$$\mathcal{H} = \sum_k \frac{1}{2} (P_k^2 + Q_k^2 - 1) \hbar\omega_k + E_0', \quad (19)$$

with

$$\hbar\omega_k = Hg\beta + 2JS[z - \sum_a \cos(\mathbf{k} \cdot \mathbf{a})], \quad (20)$$

the summation being extended over the z vectors \mathbf{a} that connect an atom with its z neighbors.

From the definitions (18) and (7) it is easily verified that the eigenvalues of the operator

$$n_k = \frac{1}{2} (P_k^2 + Q_k^2 - 1),$$

are 0, 1, 2, \dots , so that the eigenvalues of the Hamiltonian (19) are given by

$$E(n_k) = \sum_k n_k \hbar\omega_k + E_0', \quad (21)$$

in which the frequency of the normal mode \mathbf{k} is given by the dispersion law (20). We use the German letters n_k to designate the quantum numbers associated with the various spin waves of the crystal in order to distinguish them from the spin deviation operators or quantum numbers of individual atoms, which we denote by n_i , and which are not good quantum numbers when interatomic exchange coupling is included. Increase of either n_k or n_i by one unit decreases the total spin of the crystal in the z direction by unity, and the crystal's total spin deviation is given equally well by $\sum_k n_k$ or $\sum_i n_i$.

The result, (20) and (21), is the well-known expression for the spin-wave approximation to the low-lying energy levels of the spin system (1). It can be used for the evaluation of the partition function of a ferromagnetic spin system at low temperatures. However, the calculation is practical only if the temperature is so low that one may expand the right-hand member of the dispersion law (20) in powers of k and retain only the quadratic terms. If we do this, (20) becomes

$$\hbar\omega_k = Hg\beta + JS \sum_a a^2 k^2 \cos^2 \theta_{k,a}, \quad (22)$$

where $\theta_{k,a}$ is the angle between the vectors \mathbf{a} and \mathbf{k} . For any cubic array, $\cos^2 \theta_{k,a}$ may be replaced by its mean value $\frac{1}{3}$. Furthermore, for a simple, body- or face-centered lattice, one has $za^2 = 6l^2$, where l is the length of the elementary cubic cell, and then

$$\hbar\omega_k = Hg\beta + 2JSk^2 l^2. \quad (23)$$

IV. MAGNETIZATION AND SPECIFIC HEAT OF A FERROMAGNETIC IN THE SPIN-WAVE APPROXIMATION

It is customary to obtain the temperature dependence of the magnetization from (22) by setting up the partition function, and noticing that the various excitation possibilities for the spin waves correspond essentially to the Bose-Einstein type of statistics. It is, however, possible to obtain the results more simply by carrying our harmonic oscillator formalism a little further. The energy levels of a harmonic oscillator of frequency ω_k are, apart from the half-quantum of zero point energy, respectively, $\hbar\omega_k$, $2\hbar\omega_k$, \dots for the successive quantum number assignments $n_k = 1, 2, \dots$. The corresponding mean energy is well known to be

$$\bar{E}_k = \frac{\hbar\omega_k}{\exp(\hbar\omega_k/kT) - 1}. \quad (24)$$

With our spin waves, the number of spin deviations are, respectively, 1, 2, \dots for $n_k = 1, 2, \dots$. Hence the mean energy of the oscillator ω_k and the corresponding mean spin deviation differ only by a factor $\hbar\omega_k$. The total

¹⁵ For adaptation of the spin-wave theory to thin films see M. J. Klein and R. S. Smith, Phys. Rev. 81, 378 (1950).

spin deviation associated with all the different spin waves is consequently

$$\langle NS - S_z \rangle_{av} = \sum_k [\exp(\hbar\omega_k/kT) - 1]^{-1}, \quad (25)$$

where the sum is over all the allowed points of the \mathbf{k} -lattice. The deviation of the total magnetic moment M_z in the z direction from its maximum value M_{z0} achieved at $T=0$ differs from the mean spin deviation by a factor $g\beta$.

The effect of the applied field is usually small compared to the exchange coupling, so that we can take $\hbar\omega_k = 2JSk^2l^2$. If we assume that the lattice is a cubic one, and replace the sum by an integral, we get

$$M_{z0} - M_z = g\beta \left(\frac{L}{2\pi} \right)^3 \times \int \int \int \frac{dk_x dk_y dk_z}{\exp(2JS(k_x^2 + k_y^2 + k_z^2)l^2/kT) - 1}, \quad (26)$$

where L is the edge of the cubic crystal, so that $V = L^3$ is the total volume of the crystal, and where the integration runs over the first Brillouin zone in \mathbf{k} space. At low temperatures the integral may be extended over the entire \mathbf{k} space, and we then obtain

$$\begin{aligned} M_{z0} - M_z &= g\beta \left(\frac{L}{2\pi} \right)^3 4\pi \int_0^\infty \frac{k_0^2 dk_0}{\exp(2JSk_0^2l^2/kT) - 1} \\ &= \left(\frac{L}{l} \right)^3 \frac{g\beta}{2\pi^2} \left(\frac{kT}{2JS} \right)^{\frac{3}{2}} \int_0^\infty \frac{V^2}{e^{V^2} - 1} dV \\ &= \left(\frac{L}{l} \right)^3 \frac{g\beta}{2\pi^2} \left(\frac{kT}{2JS} \right)^{\frac{3}{2}} \int_0^\infty V^2 (e^{-V^2} + e^{-2V^2} + \dots) dV \\ &= \left(\frac{L}{l} \right)^3 \frac{g\beta}{2\pi^2} \left(\frac{kT}{2JS} \right)^{\frac{3}{2}} \frac{\pi^{\frac{1}{2}}}{4} \left(1 + \frac{1}{2^{\frac{3}{2}}} + \frac{1}{3^{\frac{3}{2}}} + \dots \right). \end{aligned} \quad (27)$$

The saturation magnetic moment achieved at $T=0$ is $M_{z0} = g\beta NS$. For a simple cubic lattice where $N = (L/l)^3$, our expression for M_z can be written in the form

$$\begin{aligned} M_z &= M_{z0} \left[1 - \left(\frac{kT}{2JS} \right)^{\frac{3}{2}} \left(\frac{1}{4\pi} \right)^{\frac{3}{2}} \frac{1}{S} \zeta\left(\frac{3}{2}\right) \right] \\ &= M_{z0} \left[1 - 0.1187 \left(\frac{kT}{2JS} \right)^{\frac{3}{2}} \frac{1}{S} \right]. \end{aligned} \quad (28)$$

Here and elsewhere $\zeta(m)$ denotes the Riemann zeta function $\sum_n n^{-m}$. For the body- and face-centered lattice, the coefficients of the $T^{\frac{3}{2}}$ term are, respectively, one-half and one-fourth as large as given in (28), corresponding to the fact that $(L/l)^3$ is $\frac{1}{2}N$ and $\frac{1}{4}N$ instead of N .

The expression (28) is the celebrated Bloch result² that the deviation of the spontaneous magnetic moment from its maximum value achieved at $T=0$ is proportional to $T^{\frac{3}{2}}$. Bloch considered the special case $S=\frac{1}{2}$. The extension to arbitrary S was first given by Möller.¹⁶ The $T^{\frac{3}{2}}$ law is valid for any three-dimensional lattice, as it is a consequence of the general structure of the integral (26).

An interesting result of the spin-wave theory is that it predicts no ferromagnetism for any one- or two-dimensional lattice, regardless of the number of neighbors. This is in contrast to Heisenberg's so-called Gaussian approximation,^{4,8} which made the number of nearest neighbors the only criterion, and so predicted that the simple cubic and the two-dimensional hexagonal lattice behave similarly. The absence of ferromagnetism for one- and two-dimensional lattices is a reflection of the fact that in these cases the final integral in \mathbf{k} space is in general of the type $\int [\exp(ak^2) - 1]^{-1} k^{n-1} dk$, where n is the dimensionality of the lattice. This integral diverges at the origin for $n=1$ or 2, and then the mean spin deviation is infinite. This state of affairs means that there is no stability attached to a ferromagnetic ordered state, and so ferromagnetism cannot exist if the dimensionality is less than three.

The contribution to the specific heat can be obtained by a calculation analogous to that of the magnetic moment. As the energy of a spin-wave state differs from its contribution to the deviation in magnetic moment by a factor $\hbar\omega_k/g\beta$ the intrinsic energy is given by

$$U = \left(\frac{L}{2\pi} \right)^3 4\pi \int_0^\infty \frac{(2JSk_0^2l^2)k_0^2 dk_0}{\exp(2JSk_0^2l^2/kT) - 1}, \quad (29)$$

instead of (27). Expanding in series, as in evaluating (27), we obtain

$$\begin{aligned} U &= \frac{(L/l)^3(2JS)}{2\pi^2} \left(\frac{kT}{2JS} \right)^{\frac{3}{2}} \\ &\quad \times \int_0^\infty V^4 (e^{-V^2} + e^{-2V^2} + \dots) dV, \end{aligned} \quad (30)$$

and consequently the specific heat for zero external field is given by

$$C_V = dU/dT = cNk(kT/2JS)^{\frac{3}{2}}, \quad (31)$$

where for a simple cubic lattice

$$c = \frac{5}{2} \left(\frac{1}{2\pi^2} \right) \frac{3\pi^{\frac{1}{2}}}{8} \zeta\left(\frac{5}{2}\right) = \frac{15}{8\pi^{\frac{1}{2}}} \zeta\left(\frac{5}{2}\right) = 0.113. \quad (32)$$

For body- and face-centered lattices, the formula for the constant c has an extra factor $\frac{1}{2}$ or $\frac{1}{4}$, respectively.

The exchange integral and temperature drop out of the ratio $C_V M_{z0} / (M_{z0} - M_z)$, which has the value

¹⁶ C. Möller, Z. Physik 82, 559 (1933).

0.96 $Nk/2S$ for any three-dimensional structure with cubic symmetry.

It is to be emphasized that the formulas which we have presented for the magnetization and specific heat are based on the approximations of the standard spin-wave theory. Several authors have attempted to correct them for the attractive and repulsive effects described in Sec. III, or in other words developed a more refined theory.^{12-14,17-21} The various writers do not agree in their conclusions, and so we shall not attempt to investigate this difficult and controversial subject of the higher order corrections. The most recent attempt is that of Dyson.²¹ He concludes "the practical result of the whole (his) investigation is simply this, that the linear Bloch theory with non-interacting spin waves is good enough for all practical purposes."

In the next section we discuss the corrections which result, not because of the inadequacy of the standard spin-wave theory for the Heisenberg or Heitler-London model, but rather the generalization of this model to include forces of dipolar structure.

V. INFLUENCE OF INTERACTION OF DIPOLAR STRUCTURE

We now examine how the eigenvalues associated with the spin waves are modified when dipolar coupling is superposed on isotropic exchange interaction. This problem was first posed and solved by Holstein and Primakoff.⁸ It is appreciably more complicated than the purely isotropic one considered in the preceding section. One reason is that the dipolar energy is not diagonal with respect to the components of the total crystalline spin, so that $\sum_i S_{iz}$ is not a constant of the motion.

When interaction of dipolar structure is added, the Hamiltonian function becomes

$$\mathcal{H} = -2J \sum_{\text{nei}} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{i,j} D_{ij} [\mathbf{S}_i \cdot \mathbf{S}_j - 3(\alpha_{ij} \cdot \mathbf{S}_i)(\alpha_{ij} \cdot \mathbf{S}_j)]. \quad (33)$$

Here the summation over i and j has now in general to be extended over all pairs of atoms in the crystal rather than only over nearest neighbors. The components of the unit vector α_{ij} are the direction cosines α_{ij} , β_{ij} , and γ_{ij} of the vector \mathbf{r}_{ij} that connects the atoms i and j . As mentioned in the introduction, the coefficient D_{ij} has the value $g^2\beta^2/r_{ij}^3$ if the dipolar interaction has a bona fide electromagnetic origin. Contributions to D_{ij} can also arise from anisotropic exchange, and the resulting values of D_{ij} for adjacent atoms can be appreciably greater than those given by classical

electromagnetic theory. Holstein and Primakoff utilized the classical values of D_{ij} , but the extension to the more general "pseudodipolar" case occasions no particular difficulty, and so we will not restrict D_{ij} to classical values.

The spin-wave approximation is introduced by substituting (9) in (33). The resulting terms may conveniently be grouped according to increasing powers of P_i , Q_i , and n_i , where n_i is defined by

$$n_i = \frac{1}{2}(P_i^2 + Q_i^2 - 1). \quad (34)$$

The expression (34) has the physical significance of being the spin deviation of atom i (cf. Sec. II), and can be considered of the second order compared with P_i or Q_i . When the spin wave, or harmonic oscillator approximation is made, the Hamiltonian function (33) becomes

$$\mathcal{H} = -2J \sum_{\text{nei}} \mathbf{S}_i \cdot \mathbf{S}_j + D_0 + D_1 + D_2 + D_3 + D_4, \quad (35)$$

where

$$D_0 = S^2 \sum_{i,j} D_{ij} (1 - 3\gamma_{ij}^2), \quad (36)$$

$$D_1 = -6S^4 \sum_{i,j} D_{ij} [\alpha_{ij}\gamma_{ij}Q_j + \beta_{ij}\gamma_{ij}P_j], \quad (37)$$

$$D_2 = S \sum_{i,j} D_{ij} [(1 - 3\alpha_{ij}^2)Q_iQ_j - 3\alpha_{ij}\beta_{ij}(Q_iP_j + P_iQ_j) + (1 - 3\beta_{ij}^2)P_iP_j - (1 - 3\gamma_{ij}^2)2n_i], \quad (38)$$

$$D_3 = 3S^4 \sum_{i,j} D_{ij} [\alpha_{ij}\gamma_{ij}(Q_in_j + n_iQ_j) + \beta_{ij}\gamma_{ij}(P_in_j + n_iP_j)], \quad (39)$$

$$D_4 = \sum_{i,j} D_{ij} (1 - 3\gamma_{ij}^2)n_in_j. \quad (40)$$

The terms D_3 and D_4 , which can be interpreted as anharmonic corrections, are of higher order and are henceforth neglected. We must now examine the remaining terms D_0 , D_1 , and D_2 in more detail.

The term D_0 is an additive constant as far as the calculation of the eigenvalues is concerned. It can be interpreted physically as the total dipolar, or pseudodipolar energy which results when all the spins are aligned parallel to the z axis. This is, however, an ideal condition achieved only in an infinitely strong magnetic field, for the dipolar coupling itself spoils the constancy of the magnetic moment, and so inhibits perfectly parallel alignment. In case the dipolar interaction is of the classical type, the constant D_0 may be expressed in terms of the classical demagnetization factor N_z , which is defined by

$$N_z = (V/N) \sum_{i,j} r_{ij}^{-3} (1 - 3\gamma_{ij}^2) + (4\pi/3), \quad (41)$$

with analogous definitions of N_x and N_y . Here V is the volume of the crystal, and N/V the number of spins per unit volume. The demagnetization factors have a meaning only if the demagnetizing field is uniform throughout the specimen, which is true only if the specimen is ellipsoidally cut with the external field along one of the axes; this we assume to be the case. The sum over j in (41) is then independent of i .

¹⁷ H. A. Kramers, 7e Congrès International du Froid (1936); Commun. Kamerlingh Onnes Lab., Leiden, Suppl. No. 83.

¹⁸ E. P. Wohlfarth, Proc. Leeds Phil. Lit. Soc. Sci. Sect. 5, 213 (1949).

¹⁹ W. Opechowski, Physica 4, 765 (1937).

²⁰ J. Van Kranendonk, Physica 21, 81, 749, and 925 (1955).

²¹ F. J. Dyson, Phys. Rev. 102, 1217 and 1230 (1956).

When we introduce the saturation magnetization

$$M_0 = (N/V)g\beta S, \quad (42)$$

and make use of (41), we can write the purely classical part of (36) in the form

$$D_0 = -\frac{1}{2}VM_0[(4\pi/3)M_0 - N_zM_0]. \quad (43)$$

The factor in parenthesis is equal to the sum of the Lorentz field $(4\pi/3)M_0$ and the demagnetizing field $-N_zM_0$, and is thus equal to the so-called effective field, which is the field at the position of a lattice site. It is seen from (43) that D_0 represents the energy of the magnetic moments in this field. The factor $\frac{1}{2}$ appears because the field is a result of the moments themselves so that (43) is a self-energy. The result (43) verifies the correctness of the expression (41) for the demagnetizing factor.

The term D_1 , which is given in (37) and is linear in Q_i and P_i , can be neglected for all practical purposes, for by introducing the running waves (17), one can show that the order of magnitude of D_1 is a factor $N^{\frac{1}{2}}$ smaller than that of D_0 and D_2 . Usually D_1 is even smaller, of order $N^{\frac{1}{2}}$, i.e., proportional to the number of atoms near the surface of the crystal. For the short-range forces this is true if there are symmetry planes at each atom. For then

$$\sum_j D_{ij} \alpha_{ij} \gamma_{ij} = \sum_j D_{ij} \beta_{ij} \gamma_{ij} = 0, \quad (44)$$

except for the atoms near the surface. For the long-range magnetic forces, (44) holds only if the crystal is suitably cut.

The quadratic term D_2 is the most significant of the dipolar terms. It cannot be simplified with the help of relations such as (44). Our task is now to determine the eigenvalues of

$$-2J \sum_{\text{nei}} \mathbf{S}_i \cdot \mathbf{S}_j + D_2. \quad (45)$$

The first step is to introduce the running waves (17) into the expression (38) for D_2 . The first term of D_2 then becomes equal to

$$\frac{1}{2}N^{-1}S \sum_{i,j} D_{ij} (1 - 3\alpha_{ij}^2) \sum_{k,k'} Q_k Q_{k'} \times \exp[-i\mathbf{k} \cdot \mathbf{r}_i - i\mathbf{k}' \cdot \mathbf{r}_j], \quad (46)$$

where the sum now runs over all i and j rather than over $i < j$. The expression (46) can be written in the form

$$\frac{1}{2}N^{-1}S \sum_{k,k'} Q_k Q_{k'} \sum_i \exp[-i(\mathbf{k} + \mathbf{k}') \cdot \mathbf{r}_i] \times \sum_j D_{ij} (1 - 3\alpha_{ij}^2) \exp[-i\mathbf{k}' \cdot (\mathbf{r}_j - \mathbf{r}_i)]. \quad (47)$$

We now introduce the following lattice sums

$$\begin{aligned} A_{zz}(\mathbf{k}) &= S \sum_j D_{ij} (1 - 3\alpha_{ij}^2) \exp[i\mathbf{k} \cdot (\mathbf{r}_j - \mathbf{r}_i)]; \\ A_{xy}(\mathbf{k}) &= -3S \sum_j D_{ij} \alpha_{ij} \beta_{ij} \exp[i\mathbf{k} \cdot (\mathbf{r}_j - \mathbf{r}_i)]; \end{aligned} \quad (48)$$

and similarly for $A_{yy}(\mathbf{k})$, $A_{zz}(\mathbf{k})$, and $A_{yz}(\mathbf{k})$ in an obvious notation (these quantities are clearly the components of a tensor), and we make the assumption that these quantities are independent of \mathbf{r}_i . For the

short-range forces this is justified if we neglect surface effects. For the long-range magnetic forces, however, the situation is more complicated. Sums of the type (48) with D_{ij} proportional to r_{ij}^{-3} have been discussed by Heller and Marcus²² in connection with exciton theory, and by Cohen and Keffer.²³ For $\mathbf{k}=0$, the sums (48) are related directly to the demagnetization factors (41), and for ellipsoidal crystals they are then independent of \mathbf{r}_i . For $\mathbf{k} \neq 0$, however, the quantities (48) are independent of \mathbf{r}_i only for atoms i that are away from the surface of the crystal at a distance large compared to the wavelength, since the contributions of the terms j for which r_{ij} is large compared to the wavelength cancel because of the rapidly varying phase factors. Thus for wavelengths that are small compared to the dimensions of the crystal, the sums (48) are independent of \mathbf{r}_i for the majority of the atoms i also for the long-range forces. It is only for wavelengths comparable to the dimensions of the crystal that our assumption breaks down. If we neglect the influence of these effects, the summation over i in (47) can be carried out immediately in virtue of the property

$$N^{-1} \sum_i \exp(i\mathbf{k} \cdot \mathbf{r}_i) = \delta(\mathbf{k}), \quad (49)$$

where \mathbf{k} is any of the allowed vectors in the first Brillouin zone. The expression (47) then reduces to

$$\frac{1}{2} \sum_k A_{zz}(\mathbf{k}) Q_k Q_{-k}. \quad (50)$$

The other terms in D_2 can be evaluated in the same way, and the resulting expression for D_2 is

$$\begin{aligned} D_2 &= \frac{1}{2} \sum_k [A_{xx}(\mathbf{k}) - A_{zz}(0)] Q_k Q_{-k} \\ &\quad + \frac{1}{2} \sum_k [A_{yy}(\mathbf{k}) - A_{zz}(0)] P_k P_{-k} \\ &\quad + \sum_k A_{xy}(\mathbf{k}) Q_k P_k + \frac{1}{2} N A_{zz}(0). \end{aligned} \quad (51)$$

For crystals of the bravais type, all the quantities (48) are real. It is then convenient to use the real standing waves (18) rather than the running waves (17). In terms of these quantities the expression for the Hamiltonian (35) (with D_1 , D_3 , and D_4 neglected) becomes

$$\mathcal{H} = \frac{1}{2} \sum_k [A(\mathbf{k}) Q_k^2 + B(\mathbf{k}) P_k^2 + 2C(\mathbf{k}) Q_k P_k] + E_0''. \quad (52)$$

The coefficients A , B , and C are given by

$$A(\mathbf{k}) = \hbar\omega_k^{(0)} + A_{zz}(\mathbf{k}) - A_{zz}(0); \quad (53)$$

$$B(\mathbf{k}) = \hbar\omega_k^{(0)} + A_{yy}(\mathbf{k}) - A_{zz}(0); \quad (54)$$

$$C(\mathbf{k}) = A_{xy}(\mathbf{k}); \quad (55)$$

where $\hbar\omega_k^{(0)}$ is equal to the energy (20) of a spin-wave \mathbf{k} in the absence of dipolar interaction. The constant E_0'' is

$$E_0'' = E_0' + \frac{1}{2}N(S+1)A_{zz}(0) - \sum_k \frac{1}{2}\hbar\omega_k^{(0)}, \quad (56)$$

where E_0' is defined in (14).

²² W. R. Heller and A. Marcus, Phys. Rev. **84**, 809 (1951).

²³ M. H. Cohen and F. Keffer, Phys. Rev. **99**, 1128 and 1135 (1955).

Diagonalization of the Hamiltonian (52) is easily accomplished by transforming the expression in square brackets to a simple sum of squares. This can be achieved by means of an orthogonal transformation to a new set of variables Q_k' and P_k' , which we shall not write down. The result is

$$\mathcal{H} = \sum_k \frac{1}{2} (Q_k'^2 + P_k'^2) (AB - C^2)^{\frac{1}{2}} + E_0''. \quad (57)$$

Because of the fact that the transformation is orthogonal, the eigenvalues of the operator $\frac{1}{2} (Q_k'^2 + P_k'^2)$ are again of the form $n_k + \frac{1}{2}$. The eigenvalues of the Hamiltonian (57) are therefore given by

$$E(n_k) = \sum_k n_k \cdot \hbar \omega_k + E_0, \quad (n_k = 0, 1, 2, \dots), \quad (58)$$

where the frequency of the normal mode \mathbf{k} is now

$$\hbar \omega_k = [A(\mathbf{k})B(\mathbf{k}) - C(\mathbf{k})^2]^{\frac{1}{2}}, \quad (59)$$

and where the total zero-point energy is equal to

$$E_0 = E_0'' + \frac{1}{2} \sum_k \hbar \omega_k, \quad (60)$$

E_0'' being given by (56).

The result (58) shows that in the presence of the dipolar interaction the spin system may in first approximation again be described by a set of harmonic oscillators, the normal modes of which are now specified by (59). The fact that the result (59), unlike that Sec. III, involves a radical sign is a reflection of the presence of the cross term $Q_k P_k$ in the Hamiltonian (52), which is absent if there is only isotropic exchange coupling [cf. Eq. (19)]. It can also be regarded as a reflection of the fact that $S_z' = \sum_i S_{iz}$ is no longer a constant of the motion.

The question now arises: what are the rather complicated formulas for the eigenvalues good for? Holstein and Primakoff³ made their calculation primarily to see how much at $T=0$ the dipolar coupling reduced the saturation magnetization from that corresponding to perfectly parallel alignment. When numerical values of the various constants are substituted, and the mean total spin deviation evaluated numerically, it turns out that the actual reduction in the saturation moment arising from this cause is not important, and that the Bloch relation $M_0 - M = AT^{\frac{1}{2}}$ is not appreciably affected. Holstein and Primakoff³ also studied the intrinsic susceptibility $(\partial M / \partial H)$, in other words how the moment is influenced by field strength after the directions of easy magnetization have already been rotated into alignment parallel to the applied field. The calculations³ are rather intricate. They find that for a large range of field strengths $(\partial M / \partial H) \propto T/H^{\frac{1}{2}}$. This result appears to be in accord with the limited amount of experimental evidence available.

Another physical quantity which can be studied with the aid of spin waves is ferromagnetic anisotropy near the absolute zero. This anisotropy can be caused by pseudodipolar coupling for arbitrary S , or to quadrupolar coupling, such as we mentioned in Sec.

I, for $S \geq 1$. In nickel the experimental evidence indicates that $S = \frac{1}{2}$; hence the pseudodipolar mechanism is presumably responsible for the anisotropy in this material. (Purely classical dipolar coupling would give some anisotropy, but the numerical value is insufficient.) At $T=0$, the anisotropy results from the fact that the zero-point energy (60) depends on direction. The amount and form of this directional dependence have been obtained by Tessman.²⁴ With certain simplifications, the results which he obtains are precisely the same as Van Vleck²⁵ obtained in 1937 with a semiphenomenological model based on the Weiss molecular field. At first sight the agreement of the results seems quite remarkable, as the method of approach is quite different. On tracing through the calculations, however, the accord is to be expected, being a consequence of the fact that in either calculation the change in the energy for $\Delta S_z = \mp 1$ at $T=0$ is taken to be $\pm 2Jz$ where J is the exchange integral and z is the number of nearest neighbors. Thus the "frequency denominators" in the perturbation calculations are the same, and when the denominator is independent of the transition, the completeness relation, or theorem of spectroscopic stability concerning the invariance of the sum of squares of matrix elements assures that the results will be similar. Ferromagnetic anisotropy with the dipolar model was also previously examined by Van Peipe²⁶ in a rather intricate calculation with essentially the spin-wave model and less drastic assumptions. The results do not differ greatly from those obtained by Tessman or Van Vleck.

The calculation of the anisotropy resulting from quadrupolar interaction is simpler than that for dipolar, as it can be deduced from a first rather than from a second-order perturbation calculation. The usual theories utilize some sort of a molecular field model. However, it is also possible to attack the problem with spin waves for isotropic exchange coupling as the unperturbed system on which the quadrupolar terms are superposed as a small perturbation. This approach has been utilized in an interesting paper by Keffer.²⁷ The most striking result is that the anisotropy should vary as the tenth power of the magnetization, in accord with experiments in iron.

One of the most interesting applications of the dipolar spin-wave theory is to ferromagnetic resonance, discussed in the next section.

VI. SPIN-WAVE THEORY OF FERROMAGNETIC RESONANCE ABSORPTION

In a ferromagnetic resonance absorption experiment, a small oscillating magnetic field $H_x = H_1 \cos \omega t$ is applied perpendicularly to the constant external field H , supposed to be along the z direction. As first

²⁴ J. R. Tessman, Phys. Rev. **96**, 1192 (1954).

²⁵ J. H. Van Vleck, Phys. Rev. **52**, 1195 (1937).

²⁶ W. Van Peipe, Physica **5**, 465 (1938).

²⁷ F. Keffer, Phys. Rev. **100**, 1692 (1955).

observed by Griffith,²⁸ a sharp resonance occurs in the resulting absorption of energy from the oscillating field. The expression for the resonance frequency is

$$\hbar\omega = g\beta\{[H + (N_x - N_z)M][H + (N_y - N_z)M]\}^{\frac{1}{2}}. \quad (61)$$

Here M is the magnetization of the crystal, corresponding to the external field H and temperature T . The quantities N_x , N_y , and N_z are the classical demagnetization factors, for which the microscopic definition is given in Eq. (41). The simple expression (61) is valid only for an ellipsoidal crystal with axes along the x , y , and z directions, and also only if the anisotropy is neglected.

The expression (61) was first derived by Kittel²⁹ with classical theory. A quantum-mechanical derivation was given by Van Vleck.³⁰ Both derivations are based on deductions from the equations of motion, and do not utilize spin waves. The present article derives (61) by the method of spin waves, although this is not the easiest way to derive (61). At first sight it appears that there is a sharp contradiction between Kittel's result, and the spin-wave model, for the latter has $3N$ oscillators or normal modes, where N is the number of magnetic atoms in the crystal. Most of these proper vibration frequencies are distinct, and so one might expect practically a continuum of resonance frequencies rather than only one line as predicted by (61). The answer to this riddle, as we show below, is that out of the practically infinite number of spin-wave frequencies, only one is normally "active" in magnetic resonance absorption, i.e., is allowed by the selection principles for magnetic dipole radiation. This particular frequency is the one which is symmetric in all the spins of the different atoms, and corresponds to zero propagation vector \mathbf{k} . Essentially this point was made in a derivation of (61) given by Polder,³¹ and it is our purpose to derive the same result by means of our harmonic oscillator model. It should be noted that (61) does not involve the exchange integral at all, unlike the general spin-wave frequency. Without the demagnetization corrections, the frequency (61) reduces to the Larmor frequency $g\beta H$, which is the same as the spin-wave frequency of Sec. III for the particular case $\mathbf{k}=0$ [cf. Eq. (20)]. The nonappearance of the exchange frequency in ferromagnetic resonance is to be expected, for the total dipole moment $S_z' = \sum_i S_{iz}$ commutes in matrix multiplications with the exchange energy $-2J\sum_{\text{nei}} \mathbf{S}_i \cdot \mathbf{S}_j$.

To derive an expression for the resonance frequency on the basis of the spin-wave theory, we assume that the skin depth of the oscillating field H_x is large compared to the dimensions of the crystal. The field H_x may then be considered to be uniform throughout

the crystal, and the interaction of the spin system with the oscillating field is then given by

$$\mathcal{H}' = -H_x g\beta \sum_i S_{ix}, \quad (62)$$

where the sum runs over all the atoms in the crystal. In the spin-wave approximation, the operator S_{ix} is replaced by the harmonic oscillator coordinate $S^{\frac{1}{2}}Q_i$, cf. Eq. (9), and \mathcal{H}' then becomes equal to

$$\mathcal{H}' = -H_x g\beta S^{\frac{1}{2}} \sum_i Q_i = -H_x g\beta (NS)^{\frac{1}{2}} Q_0, \quad (63)$$

where according to Eq. (17), Q_0 is the completely symmetrical normal coordinate $N^{-\frac{1}{2}} \sum_i Q_i$. Finally, we have to carry out the orthogonal transformation to the variables Q_k' and P_k' , which are the normal coordinates in the presence of dipolar interaction, cf. Eq. (57). Now Q_k' and P_k' depend only on the Q_k and P_k which have the same wave-number vector \mathbf{k} , and Q_0 is therefore a linear function only of Q_0' and P_0' . Consequently \mathcal{H}' is of the form

$$\mathcal{H}' = -H_x g\beta (NS)^{\frac{1}{2}} (aQ_0' + bP_0'), \quad (64)$$

where a and b are functions of the quantities (53), (54), and (55), the precise form of which is irrelevant at present.

The energy eigenstates of the spin system are characterized by the quantum numbers n_k which indicate the degree of excitation of the various normal modes or, equivalently, the number of excited spin waves with wave-number vector \mathbf{k} . Because of the fact \mathcal{H}' contains only the variables Q_0' and P_0' of the zeroth normal mode, the matrix elements of \mathcal{H}' in the n_k representation, between the states n_0, \dots, n_k, \dots and n_0', \dots, n_k', \dots are different from zero only if

$$n_0' = n_0 \pm 1 \quad \text{and} \quad n_k' = n_k \quad \text{for} \quad k \neq 0. \quad (65)$$

Thus the oscillating magnetic field can excite only the zeroth normal mode, i.e., only spin waves of zero wave number. The frequency of this normal mode,

$$\hbar\omega_0 = [A(0)B(0) - C(0)^2]^{\frac{1}{2}}, \quad (66)$$

is evidently equal to the resonance frequency. When we substitute the expressions (53), (54), and (55) for $A(0)$, $B(0)$, and $C(0)$, the expression (66) for the resonance frequency becomes

$$\hbar\omega_0 = \{[g\beta H + A_{zz}(0) - A_{zz}(0)] \times [g\beta H + A_{yy}(0) - A_{zz}(0)] - A_{zy}(0)^2\}^{\frac{1}{2}}. \quad (67)$$

Let us first assume that only purely classical dipolar coupling is present. The expression (67) then reduces to Kittel's formula (61). According to (41) and (48) we have

$$\begin{aligned} A_{zz}(0) &= S \sum_j g^2 \beta^2 r_{ij}^{-3} (1 - 3\gamma_{ij}^2) \\ &= g\beta N_z (N/V) g\beta S = g\beta N_z M_0, \end{aligned} \quad (68)$$

and similarly for $A_{xx}(0)$ and $A_{yy}(0)$, while $A_{zy}(0)$ may be assumed to vanish. When these results are sub-

²⁸ J. H. E. Griffith, *Nature* **138**, 670 (1946).

²⁹ C. Kittel, *Phys. Rev.* **71**, 270 (1947); **73**, 155 (1948); *J. phys. radium* **12**, 149 (1951).

³⁰ J. H. Van Vleck, *Phys. Rev.* **78**, 266 (1950); *Physica* **17**, 234 (1951).

³¹ D. Polder, *Phil. Mag.* **11**, 99 (1949).

stituted into Eq. (67), we get for the resonance frequency

$$\hbar\omega_0 = g\beta\{[H + (N_x - N_z)M_0][H + (N_y - N_z)M_0]\}^{\frac{1}{2}}, \quad (69)$$

which is identical with Kittel's expression (61), except that the magnetization M is replaced here by the saturation magnetization M_0 . Thus, as was to be expected, the spin-wave theory gives only the low-temperature value of the resonance frequency.

Let us now consider the short-range dipolar forces. These have no influence on the resonance frequency if the lattice has cubical symmetry. All the quantities $A_{xx}(0)$, etc. then vanish identically (apart from surface effects which ordinarily are negligible) because they would give rise to a quadratic dependence on the direction cosines of the external field relative to the crystal axes, and such a quadratic dependence is incompatible with cubic symmetry. The only way the short-range forces can influence the resonance frequency is through the higher order perturbations of the anharmonic terms D_3 and D_4 which we neglected. If, however, there is no cubic symmetry, the situation is quite different. The contribution of the short-range forces to the quantities $A_{xx}(0)$, etc., then does not vanish, and the anisotropy arising from these forces then has a direct effect on the resonance frequency. The same is true for the anisotropy arising from the quadrupolar coupling between the spins, as has been discussed by Van Vleck.³⁰

Finally, we discuss an alternative derivation of the expression (61) for the resonance frequency, following Luttinger and Kittel.³² From the fact that the interaction (62) between the oscillating field and the spin system commutes with the square of the total spin $\mathbf{J} = \sum \mathbf{S}_i$ of the crystal (for which we previously used the symbol \mathbf{S}'), it follows that the oscillating field induces transitions only between states having the same value of the total spin quantum number J . To calculate the resonance frequency, we must therefore calculate the energy difference between states corresponding to a definite value of J of a magnetized crystal interacting with an external field and the demagnetizing field. The classical Hamiltonian describing the energy of the crystal as a function of the orientation of its magnetization \mathbf{M} of constant magnitude M is given by

$$\mathcal{H} = -VM_zH + \frac{1}{2}V(N_xM_x^2 + N_yM_y^2 + N_zM_z^2). \quad (70)$$

Luttinger and Kittel³² observed that for deriving the ferromagnetic resonance frequency, it suffices to calculate the eigenvalues of (70), rather than those of the much more complicated Hamilton function employed by Holstein and Primakoff,³ and utilized here in Sec. V. Luttinger and Kittel showed that to a certain approximation the eigenvalues of (70) are evenly spaced

with a separation given by (61). Their proof utilized an ingenious, but somewhat devious method, in which a difference equation was replaced by a differential equation, with the axis of quantization perpendicular to the external field. The proper result can also be obtained by using an axis of quantization parallel to the magnetic field, with a rather straightforward application of the harmonic oscillator approximation.

If we assume that $M^2 = M_x^2 + M_y^2 + M_z^2$ is constant, which is legitimate because M^2 commutes quantum-mechanically with (70), then, apart from an uninteresting additive constant, (70) is the same as

$$\mathcal{H} = -VM_zH + \frac{1}{2}V[(N_x - N_z)M_x^2 + (N_y - N_z)M_y^2]. \quad (71)$$

We neglect saturation effects, i.e., we assume that M_z deviates but slightly from its maximum value M . Saturation effects, such as observed by Bloembergen and Wang,³³ appear for very intense oscillating fields which we do not consider.

The quantum-mechanical operator corresponding to (71) is obtained by replacing \mathbf{M} by $g\beta\mathbf{J}/V$,

$$\mathcal{H} = -Hg\beta J_z + \frac{1}{2}(g\beta M/J) \times [(N_x - N_z)J_x^2 + (N_y - N_z)J_y^2]. \quad (72)$$

We are interested in the eigenvalues of (72) for a very large value of J (of order 10^{20}) and for average values of J_x that do not deviate appreciably from J , this being the analog of the classical condition $M_x \approx M$. [If $N_x \neq N_y$, J_z does not commute with (72), and does not correspond to a good quantum number; hence we must then deal with the expectation or average value rather than with the eigenvalue of J_z .] But these are precisely the conditions under which the harmonic oscillator approximation (7), viz.,

$$J_x = J^{\frac{1}{2}}Q; \quad J_y = J^{\frac{1}{2}}P; \quad J_z = J - \frac{1}{2}(P^2 + Q^2 - 1), \quad (73)$$

is an extremely good approximation. When we substitute (73) into (72), we get

$$\mathcal{H} = \text{const} + g\beta^{\frac{1}{2}}(aQ^2 + bP^2), \quad (74)$$

where $a = H + (N_x - N_z)M$ and $b = H + (N_y - N_z)M$. By carrying out the simple transformation,

$$Q = (b/a)^{\frac{1}{2}}Q', \quad P = (a/b)^{\frac{1}{2}}P', \quad (75)$$

the Hamiltonian becomes

$$\mathcal{H} = \text{const} + g\beta(ab)^{\frac{1}{2}}(P'^2 + Q'^2), \quad (76)$$

from which it follows immediately that the eigenvalues of H for which $M_x \approx M$ are uniformly spaced with an energy difference $g\beta(ab)^{\frac{1}{2}}$. Substituting the values of a and b , this reduces exactly to the expression (61) for the resonance frequency.

From this derivation as well as from the analogous classical derivation, it is clear that for the higher energy levels, from which the average value of J_z differs

³² J. M. Luttinger and C. Kittel, *Helv. Phys. Acta* **21**, 480 (1948).

³³ N. Bloembergen and S. Wang, *Phys. Rev.* **93**, 72 (1954).

appreciably from J , deviations will appear from the uniform spacing (61), since for these states the harmonic oscillator approximation (73) is no longer a good approximation.

In closing this section, it is well to give a word of caution that it is becoming increasingly clear experimentally that often the phenomena of ferromagnetic resonance are much too complex to describe by the simple Kittel formula (61). Subsidiary peaks sometimes appear, especially at high power levels, where saturation effects must be considered. Because the dipolar forces fall off only as the inverse cube, boundary corrections cannot be dismissed as debonairly as is usually done, and there can be important distorting effects because of perturbations between the main mode (61) and subsidiary modes of nearly equal frequency. We do not discuss this intricate, but practically important subject, of which the literature is rapidly increasing. Particular reference should be made to papers by Clogston, Suhl, Walker, and Anderson.³⁴

VII. HARMONIC OSCILLATOR APPROXIMATION FOR ANTIFERROMAGNETISM

The starting point of a spin-wave theory of the low-temperature properties of a spin system is the ground state of the spin system: spin waves are introduced to describe small deviations of the state of the spin system from the ground state. When one wants to investigate the effect of a small perturbation, such as the application of a weak external field or of a small amount of thermal energy, the spin-wave theory is applicable only if the ground state is stable against the perturbation. In that case, the resulting small deviation of the state of the spin system from the ground state can be analyzed with the help of a set of appropriate normal modes, as explained in Sec. III for a ferromagnetic spin system. If the ground state is not stable against the perturbation, the spin-wave theory breaks down. The resulting large deviation of the state of the spin system from the initial state cannot be described in terms of the spin waves, and the results of spin-wave theory diverge for such a case.

Let us first consider the question of the stability of the ground state, and the applicability of the spin-wave theory, for the ferromagnetic case discussed in Sec. III. When we assume that there is no external field present, and no anisotropy of any kind, the Hamiltonian function of the spin system is

$$\mathcal{H} = -2J \sum_{\text{nei}} \mathbf{S}_i \cdot \mathbf{S}_j. \quad (77)$$

In the ground state all the spins are lined up parallel, but the direction of the resulting spin is arbitrary, so that the ground state is degenerate. One can, of

course, select a definite one of these states, which the spins pointing in a definite direction, as the initial state in a spin-wave theory. But with the Hamiltonian (77) this state would not be stable against small perturbations. A small amount of thermal energy, for instance, would lead to a vanishing of the total magnetic moment, corresponding to the fact that in thermal equilibrium, in the absence of any anisotropy or external field, the total magnetic moment is zero. The existence of a freely revolving total spin would give rise to divergencies in the spin-wave theory, and the theory would not be applicable. Some of the spin waves (those with $k=0$) would have zero energy, or zero frequency, and at any finite temperature these spin-wave modes would be excited with an arbitrarily large number of quanta. This difficulty can be avoided by removing the degeneracy of the ground state, either by assuming that from the outset a sufficiently large external field is present, or by introducing an effective anisotropy field H_A . In the ground state the spins are then aligned in the direction of the field. At temperatures T for which kT is small compared to $g\beta H$ or $g\beta H_A$, only small deviations from this state occur, which can be described with the help of spin waves. In reality, the anisotropy arises from some real anisotropic coupling between the spins, for example of the pseudodipolar type discussed in Sec. I. When this coupling is included in the Hamiltonian (77), the ground state becomes automatically nondegenerate, except for a trivial degeneracy resulting from the symmetry of the crystal. Unfortunately, this problem is too difficult to be handled satisfactorily, and one therefore has to introduce the artifice of an effective anisotropy field. In the ferromagnetic case the anisotropy does not play an essential role, and to remove the degeneracy of the ground state one usually introduces an external field, as in Sec. III.

Let us now consider an antiferromagnetic spin system. We restrict ourselves to the simple and body-centered cubic structures which can be divided into sublattices, 1 and 2, in such a way that all the nearest neighbors of an atom on sublattice 1 are on sublattice 2, and vice versa. This restriction is not essential, and is introduced only to keep the discussion as simple as possible. With only isotropic coupling between neighboring spins, the Hamiltonian of the spin system is given by

$$\mathcal{H} = 2J \sum_{\text{nei}} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (78)$$

where i refers to an atom on sublattice 1, and j to one on sublattice 2; the coupling constant J is positive. By an antiferromagnetic state of the two sublattice kind we shall mean a state in which there exists a certain long range order among the spin directions on sublattice 1, and a similar order among the spins on sublattice 2 in the opposite direction. With the completely isotropic Hamiltonian (78) such a state is degenerate, since the common direction of the resulting

³⁴ Clogston, Suhl, Walker, and Anderson, *Phys. Rev.* **101**, 903 (1956); *J. Phys. Chem. Solids* **1**, 129 (1956); H. Suhl, *Phys. Rev.* **101**, 1437 (1956); *Proc. Inst. Radio Engrs.* **44**, 1270 (1956); P. W. Anderson and H. Suhl, *Phys. Rev.* **100**, 1789 (1955); L. R. Walker, *Phys. Rev.* **105**, 390 (1957).

antiparallel magnetic moments of the two sublattices is arbitrary. This degeneracy cannot be removed by an external field. For a not too large external field, the spins would arrange themselves antiferromagnetically in a plane perpendicular to the external field, but the direction of the spins in the plane would still be arbitrary.

In any real antiferromagnetic crystal this degeneracy is removed by the anisotropy which, as we shall see, plays a much more important role here than in the ferromagnetic case. The anisotropy must again be introduced in the form of an effective anisotropy field which, however, is quite different from the corresponding ferromagnetic anisotropy field. It must be such that the spins on sublattice 1 are preferentially oriented in the $+z$ direction, say, and those on sublattice 2 in the $-z$ direction. This can be achieved by introducing a hypothetical field H_A pointing in the $+z$ direction at the sites of sublattice 1, and in the $-z$ direction at those of sublattice 2. The Hamiltonian then becomes

$$\mathcal{H} = 2J \sum_{\text{nei}} \mathbf{S}_i \cdot \mathbf{S}_j - H_A g\beta (\sum_i S_{iz} - \sum_j S_{jz}), \quad (79)$$

where i runs over all the atoms on sublattice 1, and j over those on sublattice 2, a convention we shall follow throughout this section. In the limit of an infinitely strong anisotropy field H_A , the coupling term in (79) may be neglected, and the ground state of (79) is then the state in which all the spins on sublattice 1 are pointing in the $+z$ direction, and those on sublattice 2 in the $-z$ direction. For a finite value of H_A the ground state of (79) turns out to be only slightly dependent on the value of H_A as long as $g\beta H_A$ is not smaller than of order J/N , where N is the total number of atoms in the crystal. More precisely, $g\beta H_A$ must be large compared to the separation in energy between the first excited state and the ground state of (78) which energy is of order J/N . (This is not true for a one-dimensional crystal. Kasteleyn³⁵ has shown that for a linear chain the ground state is not ordered when the anisotropy becomes smaller than some finite critical value. The same is probably true of a two-dimensional crystal.)

The ground state of the Hamiltonian (79) for a finite value of N , and H_A rigorously equal to zero, i.e., the ground state of the isotropic Hamiltonian (78), is not known. Fortunately, this state is not of physical interest, because it is never realized in any real antiferromagnetic crystal. The ground state that is realized for any finite value of the anisotropy is a definite linear combination of the low-lying states of (78), and it is this definite linear combination of states of (78) that we shall call the antiferromagnetic ground state. We accordingly define the antiferromagnetic ground state, corresponding to the assumed two sublattice structure, as the limiting ground state of (79) for small

H_A , or more precisely, as the state obtained when first N is made to tend to infinity, and then H_A to zero. As remarked already, and as is shown in detail below, this state does not differ much from the ground state of (79) for a finite value of H_A . In particular it does not differ much from the completely ordered state obtained by letting H_A tend to infinity. This completely ordered state is used as the initial state in the spin-wave theory of antiferromagnetism. The antiferromagnetic spin waves are introduced to describe the small deviations of the state of the spin system from this completely ordered state, which result from the fact that in reality the value of H_A is finite rather than infinite and from the application of some other small perturbation such as a weak external field or a small amount of thermal energy. If this spin-wave theory gives convergent results, the assumed antiferromagnetic ordering corresponds to at least a relative minimum in the energy. Whether this structure also corresponds to an absolute minimum cannot be decided until the problem of a spin system with a true anisotropic coupling between the spins is solved.

In the completely ordered state all spins on sublattice 1 are pointing in the $+z$ direction, and all spins on sublattice 2 in the $-z$ direction, i.e., $S_{iz}=S$ and $S_{jz}=-S$ for all i and j . If S_{iz} is not equal to S , or S_{jz} not equal to $-S$, we say that a spin deviation is present on atom i or j , respectively. The antiferromagnetic ground state already defined is then a state in which there are relatively few spin deviations present; for a simple cubic lattice it turns out that on an average about 93% of the atoms have no spin deviations. This state can therefore be treated to a fair approximation by the spin-wave theory. In general, it is not stable against small perturbations, and if one wants to investigate the effect on the state of the spin system of an external field or of the thermal energy, it is necessary to have a finite anisotropy present. The anisotropy is quite effective in insuring stability; it is not necessary that $g\beta H_A$ be large with respect to the perturbation, it is sufficient that $g\beta(H_A H_E)^{\frac{1}{2}}$ is sufficiently large, where H_E is the Weiss molecular field. The fact that the molecular field comes in here via the anisotropy is the reason why the anisotropy is so much more important here than in the ferromagnetic case. Typical values are $H_A=10^2\text{oe}$, $H_E=10^6\text{oe}$, so that, for example, an external field of the order of 10^3oe may still be regarded as a small perturbation. The effect of such small perturbations can be described satisfactorily by the spin-wave theory as long as the resulting deviations from the completely ordered state, due to the perturbation in question and the fact that H_A is not infinite, are small.

We now show how the harmonic oscillator approximation is introduced analytically in the antiferromagnetic case. When we include the interaction with a parallel external field $H_z=H$, the Hamiltonian (79) becomes

³⁵ P. W. Kasteleyn, *Physica* 18, 104 (1952).

equal to

$$\mathcal{H} = 2J \sum_{\text{nei}} \mathbf{S}_i \cdot \mathbf{S}_j - (H + H_A) g\beta \sum_i S_{iz} + (H - H_A) g\beta \sum_j S_{jz}. \quad (80)$$

In the completely ordered state we have $S_{iz} = S$ and $S_{jz} = -S$, and we therefore introduce the following spin-deviation quantum numbers

$$n_i = S - m_i \quad \text{and} \quad n_j = S + m_j. \quad (81)$$

The harmonic oscillator approximation (7) for the spin variables must be written in the following form

$$S_{ix} = S^{\frac{1}{2}} Q_i; \quad S_{iy} = S^{\frac{1}{2}} P_i; \quad S_{iz} = S - \frac{1}{2} (P_i^2 + Q_i^2 - 1); \quad (82)$$

$$S_{jx} = S^{\frac{1}{2}} Q_j; \quad S_{jy} = -S^{\frac{1}{2}} P_j; \quad S_{jz} = -S + \frac{1}{2} (P_j^2 + Q_j^2 - 1). \quad (83)$$

The matrix elements of the left-hand sides of (82) and (83) are then equal to the corresponding matrix elements of the right-hand sides for n_i and n_j equal to 0 and 1, i.e., for m_i equal to S and $S-1$, and for m_j equal to $-S$ and $-S+1$. When we substitute (82) and (83) into the Hamiltonian (80), we get

$$\mathcal{H} = E_0' + 2JS \sum_{\text{nei}} (Q_i Q_j - P_i P_j) - 2J \sum_{\text{nei}} n_i n_j + [2JSz + g\beta(H_A + H)] \sum_i n_i + [2JSz + g\beta(H_A - H)] \sum_j n_j, \quad (84)$$

where we have introduced the spin deviation operators

$$n_i = \frac{1}{2} (P_i^2 + Q_i^2 - 1) \quad \text{and} \quad n_j = \frac{1}{2} (P_j^2 + Q_j^2 - 1), \quad (85)$$

and where the quantity E_0' is equal to

$$E_0' = -NzJS^2 - H_A g\beta NS. \quad (86)$$

The properties of the system of harmonic oscillators (84) are the same as those of the spin system (80) so long as the states with $n \geq 2$ of the oscillators are not appreciably excited. For sufficiently large values of H_A this is certainly the case. However (84) is a good approximation also for small values of H_A .

In addition to the approximation (82) and (83) of replacing the spin variables by harmonic oscillator variables, we must neglect the attractive interaction between the spin deviations, which is represented by the terms of the fourth degree in (84). When we do this, the Hamiltonian can be diagonalized by introducing the appropriate normal modes. We first introduce running waves for the sublattices 1 and 2 separately,

$$\begin{cases} Q_{1k} = (2/N)^{\frac{1}{2}} \sum_i \exp(i\mathbf{k} \cdot \mathbf{r}_i) Q_i; \\ P_{1k} = (2/N)^{\frac{1}{2}} \sum_i \exp(-i\mathbf{k} \cdot \mathbf{r}_i) P_i; \end{cases} \quad (87)$$

$$\begin{cases} Q_{2k} = (2/N)^{\frac{1}{2}} \sum_j \exp(-i\mathbf{k} \cdot \mathbf{r}_j) Q_j; \\ P_{2k} = (2/N)^{\frac{1}{2}} \sum_j \exp(i\mathbf{k} \cdot \mathbf{r}_j) P_j. \end{cases} \quad (88)$$

The wave vector \mathbf{k} takes on the $\frac{1}{2}N$ different values allowed by the boundary conditions, so that we have the correct number of variables. In terms of these

variables the Hamiltonian becomes

$$\mathcal{H} = E_0' + \sum_k \{ [2JSz + g\beta(H_A + H)] \times \frac{1}{2} (P_{1k} P_{1-k} + Q_{1k} Q_{1-k} - 1) + [2JSz + g\beta(H_A - H)] \frac{1}{2} (P_{2k} P_{2-k} + Q_{2k} Q_{2-k} - 1) + 2JS\gamma_k (Q_{1k} Q_{2k} - P_{1k} P_{2k}) \}. \quad (89)$$

The quantity γ_k is given by

$$\gamma_k = z^{-1} \sum_a \cos(\mathbf{k} \cdot \mathbf{a}), \quad (90)$$

where the sum runs over the \mathbf{z} vectors \mathbf{a} that connect an atom with its z nearest neighbors. By carrying out a linear transformation (cf. references 7 and 40) of the form

$$\begin{cases} Q_{1k} = c_{1k} Q_{1k}' + c_{2k} Q_{2-k}'; \\ Q_{2-k} = c_{2k} Q_{1k}' + c_{1k} Q_{2-k}'; \\ P_{1k} = c_{1k} P_{1k}' - c_{2k} P_{2-k}', \\ P_{2-k} = -c_{2k} P_{1k}' + c_{1k} P_{2-k}', \end{cases} \quad (91)$$

where $c_{1k} = c_{1-k}$, $c_{2k} = c_{2-k}$ and $c_{1k}^2 - c_{2k}^2 = 1$ we can eliminate the cross terms in (89). When we substitute (91) into the Hamiltonian (89), we find that the cross terms vanish if the coefficients c_{ik} have the following values:

$$c_{1k} = \rho_k / (\rho_k^2 - \gamma_k^2)^{\frac{1}{2}}; \quad c_{2k} = -\gamma_k / (\rho_k^2 - \gamma_k^2)^{\frac{1}{2}}, \quad (92)$$

where γ_k is given by (90), and where ρ_k is defined as

$$\rho_k = 1 + H_A/H_E + [(1 + H_A/H_E)^2 - \gamma_k^2]^{\frac{1}{2}}. \quad (93)$$

The quantity H_E is given by

$$H_E = 2JSz/g\beta,$$

and is the Weiss molecular field in the completely ordered state. In terms of the primed variables the Hamiltonian becomes of the standard form

$$\mathcal{H} = E_0 + \sum_k \frac{1}{2} (P_{1k}' P_{1-k}' + Q_{1k}' Q_{1-k}' - 1) \hbar\omega_{1k} + \sum_k \frac{1}{2} (P_{2k}' P_{2-k}' + Q_{2k}' Q_{2-k}' - 1) \hbar\omega_{2k}. \quad (94)$$

Consequently the eigenvalues are given by

$$E(\mathbf{n}_{1k}, \mathbf{n}_{2k}) = E_0 + \sum_k (\mathbf{n}_{1k} \hbar\omega_{1k} + \mathbf{n}_{2k} \hbar\omega_{2k}), \quad (95)$$

where \mathbf{n}_{1k} and \mathbf{n}_{2k} can take the values 0, 1, 2, ... The frequencies of the normal spin-wave modes are given by

$$\begin{cases} \hbar\omega_{1k} = g\beta[(H_E + H_A)^2 - \gamma_k^2 H_E^2]^{\frac{1}{2}} + g\beta H; \\ \hbar\omega_{2k} = g\beta[(H_E + H_A)^2 - \gamma_k^2 H_E^2]^{\frac{1}{2}} - g\beta H; \end{cases} \quad (96)$$

and the energy E_0 of the ground state is equal to

$$E_0 = E_0' + \frac{1}{2} \sum_k (\hbar\omega_{1k} + \hbar\omega_{2k}), \quad (97)$$

where E_0' is given by (86).

As we see from (96), the normal modes fall into two branches which become identical for $H=0$. A detailed discussion and a classical interpretation of these spin-wave modes has been given by Keffer, Kaplan, and Yafet.⁷ Also from (96), because of the fact that $\gamma_k^2 \leq 1$,

the mode 2, 0 has the smallest energy of excitation, namely,

$$\hbar\omega_{20} = g\beta(H_c - H); \quad H_c = [H_A(H_A + 2H_E)]^{\frac{1}{2}}. \quad (98)$$

When H becomes equal to or larger than the critical value H_c , the harmonic oscillator approximation evidently breaks down, even at the absolute zero, since the mode 2, 0 would be excited to an arbitrarily high degree. This corresponds to the well-known fact that when the parallel field H exceeds the critical value H_c , the spins turn to a direction perpendicular to H . The resulting large change in the state of the spin system cannot be described by the spin-wave approximation, and this shows up in the appearance of divergencies in the theory. But the value of the critical field is given correctly by (98), at least at the absolute zero.

The case of a perpendicular external field can be treated in a similar way (cf., e.g., Kubo³⁶). The results (96) and (97) and the corresponding results for the perpendicular case can be used to calculate the parallel and the perpendicular susceptibility. The specific heat of an antiferromagnetic is discussed in connection with ferrimagnetism in Sec. IX.

As regards antiferromagnetic resonance absorption, one can show in the same way as in Sec. VI for the ferromagnetic case that an oscillating magnetic field can excite only the spin-wave modes with $\mathbf{k}=0$. From (96) we see that for a parallel external field the resonance frequencies are given by

$$\begin{cases} \hbar\omega_1 = g\beta[H_A(H_A + 2H_E)]^{\frac{1}{2}} + g\beta H; \\ \hbar\omega_2 = g\beta[H_A(H_A + 2H_E)]^{\frac{1}{2}} - g\beta H; \end{cases} \quad (99)$$

showing that the anisotropy plays an important role in the resonance phenomena. The frequencies for the perpendicular case can be derived in a similar manner. Further details of the spin-wave theory of antiferromagnetic resonance further are found in the literature.^{7,37-41} Formula (99) for the antiferromagnetic resonance frequencies was first obtained by Kittel.⁴²

VIII. ANTIFERROMAGNETIC GROUND STATE IN THE SPIN-WAVE APPROXIMATION

We shall now discuss, for our two-sublattice model, the antiferromagnetic ground state as defined in Sec. VII. This is the lowest state of the Hamiltonian (79) which represents an isotropic interaction between neighboring spins and an anisotropy energy of the spins in a staggered anisotropy field H_A corresponding to the postulated two sublattice structure. We show in particular that this ground state of (79) for $H_A \rightarrow 0$

does not differ much from the completely ordered state realized for $H_A \rightarrow \infty$, in which all the spins on sublattice 1 are pointing N , say, and those on sublattice 2 are pointing S .

In the spin-wave approximation, the Hamiltonian (79) is replaced by a system of harmonic oscillators described by the coordinates and momenta Q_i, P_i given by (82) and (83). In Sec. VII a set of approximate normal modes for this system of coupled oscillators was found given by the transformations (87), (88), and (91). In terms of these variables, if we neglect the terms of the fourth degree in the Hamiltonian (84) of the system of oscillators, the Hamiltonian becomes of the standard form (94). In the absence of an external field, the frequencies of the normal modes are all positive, as can be seen from (96) by putting $H=0$. The ground state of the oscillator system is then characterized by the fact that none of the normal modes is excited, i.e., that all n_{1k} and n_{2k} vanish. This does not mean, however, that all the original, decoupled oscillators that replace the individual spins of the spin system are necessarily in their lowest state. This is true only for $H_A \rightarrow \infty$, corresponding to the completely ordered state. For a finite value of H_A , and in particular for $H_A \rightarrow 0$, however, there exists a certain nonvanishing probability p_n that a given oscillator is in its n th excited state, which we shall calculate presently. As long as p_n for $n \geq 1$ is small compared to unity, p_n for $n=0, 1, \dots, 2S$ is approximately equal to the probability to find n spin deviations on a given atom in the ground state of the original spin system, since under the conditions $p_n \ll 1$ for $n \geq 1$ the properties of the oscillator system are approximately equal to those of the spin system. In the oscillator system the p_n have, of course, a meaning for all n .

For the calculation of the probabilities p_n as functions of the anisotropy field H_A , it is convenient to introduce creation and annihilation operators. For the spin-wave modes (91) these operators are defined as

$$\begin{aligned} A_{1k} &= 2^{-\frac{1}{2}}(Q_{1k}' + iP_{1-k}'); \\ A_{1k}^* &= 2^{-\frac{1}{2}}(Q_{1-k}' - iP_{1k}'), \end{aligned} \quad (100)$$

and similarly for the modes $2k$. The spin-deviation operator n_i corresponding to the number of spin deviations on atom i can be expressed in terms of the operators (100) by means of the transformations (87), (88), and (91). The result is

$$n_i = (2/N) \sum_{k, k'} \exp[i(\mathbf{k}' - \mathbf{k}) \cdot \mathbf{r}_i] B_{k'}^* B_k, \quad (101)$$

where

$$B_k = c_{1k} A_{1k} + c_{2k} A_{2k}^*. \quad (102)$$

The expectation value of n_i in the ground state ψ_0 which is characterized by the relations,

$$A_{1k}\psi_0 = 0, \quad A_{2k}\psi_0 = 0,$$

is independent of i and is therefore equal to

$$\langle n_i \rangle = (2/N) \sum_i \langle n_i \rangle = (2/N) \sum_k \langle B_k^* B_k \rangle, \quad (103)$$

³⁶ R. Kubo, Phys. Rev. 87, 568 (1952); Revs. Modern Phys. 25, 344 (1953).

³⁷ L. Hulthen, Proc. Acad. Sci. Amsterdam 39, 190 (1936).

³⁸ P. W. Anderson, Phys. 86, 694 (1952).

³⁹ Nagamiya, Yoshida, and Kubo, *Advances in Physics* (Taylor and Francis, Ltd., London, 1955), Vol. 4, p. 1.

⁴⁰ T. Nakamura, Progr. Theoret. Phys. 7, 539 (1952).

⁴¹ J. K. Tessman, Phys. Rev. 88, 1132 (1952).

⁴² C. Kittel, Phys. Rev. 82, 565 (1951); F. Keffer and C. Kittel, Phys. Rev. 85, 329 (1952).

where the sum over i runs over all the atoms belonging to the same sublattice as i . Only one term in $B_k^* B_k$ gives a nonvanishing contribution to the expectation value, *viz.*, $A_{2k} A_{2k}^*$, and we therefore get

$$\langle n_i \rangle = (2/N) \sum_k c_{2k}^2 = \Gamma(1 + H_A/H_B), \quad (104)$$

where the function $\Gamma(x)$ is defined as

$$\Gamma(x) = (2/N) \sum_k \frac{1}{2} [(1 - \gamma_k^2/x)^{-1/2} - 1], \quad (105)$$

and γ_k is given by (90).

In a similar way one can evaluate the expectation values of the higher powers of n_i . For n_i^2 one obtains

$$\langle n_i^2 \rangle = (2/N)^2 \sum_{k,k'} \sum_{l,l'} \delta(k+l-k'-l') \times \langle B_k^* B_k B_{l'}^* B_{l'} \rangle. \quad (106)$$

There are now two nonvanishing terms, and the result is

$$\langle n_i^2 \rangle = \Gamma + 2\Gamma^2, \quad (107)$$

where use has been made of the relation $c_{1k}^2 = 1 + c_{2k}^2$. Finally, we have calculated $\langle n_i^3 \rangle$. There are six nonvanishing terms now, giving,

$$\langle n_i^3 \rangle = \Gamma + 6\Gamma^2 + 5\Gamma^3. \quad (108)$$

The expectation values $\langle n_i^m \rangle$ are related to the probabilities p_n by the equation

$$\langle n_i^m \rangle = p_1 + 2^m p_2 + 3^m p_3 + \dots, \quad (109)$$

and we thus get the following set of equations for the p_n :

$$\begin{cases} p_0 + p_1 + p_2 + p_3 + \dots = 1; \\ p_1 + 2p_2 + 3p_3 + \dots = \Gamma; \\ p_1 + 4p_2 + 9p_3 + \dots = \Gamma + 2\Gamma^2; \\ p_1 + 8p_2 + 27p_3 + \dots = \Gamma + 6\Gamma^2 + 5\Gamma^3. \\ \dots \end{cases} \quad (110)$$

These equations can be solved by a successive approximation method by considering Γ as a small quantity. To the third order in Γ we thus get

$$\begin{cases} p_0 = 1 - \Gamma + \Gamma^2 - (5/6)\Gamma^3; \\ p_1 = \Gamma - 2\Gamma^2 + (5/2)\Gamma^3; \\ p_2 = \Gamma^2 - (5/2)\Gamma^3; \\ p_3 = (5/6)\Gamma^3. \end{cases} \quad (111)$$

From (105) we see that for $H_A \rightarrow \infty$ we have $\Gamma \rightarrow 0$ and hence $p_0 = 1$, $p_1 = p_2 = \dots = 0$, verifying the result that for very large anisotropy the ground state approaches to the completely ordered state. For $H_A \rightarrow 0$, Γ tends to a finite value which can be expressed as follows in terms of the integral J_D evaluated by Anderson³⁸:

$$\Gamma(1) = \frac{1}{2}(J_D - 1),$$

where D is the dimensionality of the lattice. For the 3-dimensional simple cubic lattice we thus obtain $\Gamma(1) = 0.078$, corresponding to the following values of the p_n :

$$p_0 = 0.93; \quad p_1 = 0.06; \quad p_2 = 0.01. \quad (112)$$

From these values it is evident that the ground state for $H_A \rightarrow 0$ does not differ much from the completely ordered state, and this result justifies the use of the spin-wave approximation for the antiferromagnetic ground state, even in the case $S = \frac{1}{2}$ for which the approximation is worst.

The values of the p_n given by (112) are independent of the spin quantum number S , since the quantity $\Gamma(1)$ depends only on the lattice structure. As only the first few p_n are appreciably different from zero, it follows that in the classical limit $S \rightarrow \infty$, the spins are completely aligned even in the absence of the anisotropy. This should be so, of course, since it is well known that with classical spin vectors the completely ordered state is the lowest energy state. For a finite value of S , the energy of the ground state can be calculated from (96) and (97). For further details we refer to the papers of Anderson³⁸ and Kubo.³⁶

Finally, the great service of the spin-wave theory in connection with antiferromagnetism is to show that there is some real foundation for the semiphenomenological model used by the various writers, wherein the spins on one sublattice are pointing N and those on the other sublattice S . The result (112) shows, so to speak, that this model is only about 7% wrong. The experiments of Shull and Smart⁴³ on neutron diffraction showed unequivocally that the model of interlocking sublattices has physical reality. Nevertheless it is reassuring that the same arrangement is predicted by theory. The worry was that the interlocking structure might be completely washed away by the perturbations nondiagonal in the total spin of either sublattice, though diagonal in their sum. The objection may also be raised that there is no way of determining which sublattice points N and which S , rather than vice versa. Actually there is a quantum-mechanical resonance effect whereby the two alignments are interchanged with each other. This, however, is a process which is almost of geological magnitude in slowness for any crystal of macroscopic size, as the resonance comes in only as a result of perturbation theory carried to the order N of atoms in the lattice. So the assignment of one type of spin to one sublattice and one to the other can be regarded as stable for all intents and purposes, like isomeric forms of heavy molecules.

IX. HARMONIC OSCILLATOR APPROXIMATION FOR FERRIMAGNETISM

One of the outstanding developments of the last decade in magnetism has been the development and fruition of the concept of ferrimagnetism. The idea of a ferrimagnetic medium is one introduced by Néel⁴⁴;

⁴³ C. G. Shull and J. S. Smart, Phys. Rev. **76**, 1256 (1949); Shull, Wollan, and Strausser, Phys. Rev. **83**, 333 (1951); C. G. Shull and M. K. Wilkinson, Revs. Modern Phys. **25**, 100 (1953).

⁴⁴ L. Néel, Ann. phys. **3**, 137 (1948). The magnetic implications of the ferrites are reviewed very completely in a special issue of Proc. Inst. Radio Engrs. (October, 1956).

it differs from an antiferromagnetic material in that the moments of the two sublattices are unequal, be it the result of unequal spins, g factors, or number of sites, so that the resultant magnetic moment of the crystal is not zero even when the two sublattices are aligned antiparallel. The spin-wave theory of the preceding sections can easily be adopted to ferrimagnetic media in which the two sublattices have equal number of atoms, and in which the only important coupling is between adjacent atoms located on different sublattices. Actual ferrimagnetic media often involve more complicated geometries as well as interactions between atoms which are not nearest neighbors. The spin-wave theory for the case that there is also a coupling between the spins on one sublattice has been given by Kaplan.⁴⁶ With the simplified model, however, the calculations of the preceding sections can be taken over with only minor modifications.

In contradistinction to the case of a true antiferromagnetic, we can no longer assume that the two sublattices have the same g factors, spins or anisotropy fields. In place of Eqs. (80) and (81), we now have

$$\mathcal{H} = 2J \sum_{\text{nei}} \mathbf{S}_i \cdot \mathbf{S}_j - (H + H_{A1}) g_1 \beta \sum_i S_{iz} - (H - H_{A2}) g_2 \beta \sum_j S_{jz} \quad (113)$$

and

$$n_i = S_1 - m_i, \quad n_j = S_2 + m_j. \quad (114)$$

We replace S by S_1 and S_2 , respectively, in the expression (82) and (83) and introduce the appropriate harmonic oscillator variables in place of the spin variables. Instead of (84), the Hamiltonian function of the oscillator system is now

$$\mathcal{H} = E_0'' + 2J(S_1 S_2)^{\frac{1}{2}} \sum_{\text{nei}} (Q_i Q_j - P_i P_j) - 2J \sum_{\text{nei}} n_i n_j + [2J S_2 z + g_1 \beta (H + H_{A1})] \sum_i n_i + [2J S_1 z + g_2 \beta (H - H_{A2})] \sum_j n_j, \quad (115)$$

where

$$E_0'' = -N z J S_1 S_2 - N(g_1 S_1 - g_2 S_2) \beta H + -N g_1 S_1 \beta H_{A1} - N g_2 S_2 \beta H_{A2}. \quad (116)$$

The expression (115) has precisely the same structure as (84); the only difference is that the coefficients are different, and that the constant (116) now contains a term proportional to the external field. Hence the Hamiltonian function (115) can be reduced to a sum of squares by essentially the same type of transformation as was utilized in Sec. VII. When allowance is made for the new values of the constants, the expressions for the frequencies of the normal modes become

$$\begin{aligned} \hbar \omega_{1k}, \hbar \omega_{2k} &= \{ [J(S_1 + S_2)z + (K_1 + K_2) + \frac{1}{2}(g_1 - g_2)\beta H]^2 \\ &\quad - 4J^2 z^2 S_1 S_2 \gamma_k^2 \}^{\frac{1}{2}} \pm [J(S_2 - S_1)z \\ &\quad + \frac{1}{2}(g_1 + g_2)\beta H + (K_1 - K_2)], \quad (117) \end{aligned}$$

instead of (96), where the $+$ sign corresponds to ω_{1k} and the $-$ sign to ω_{2k} . Here γ_k is defined as in (90).

⁴⁶ H. Kaplan, Phys. Rev. **86**, 121 (1952).

Following Kouvel and Brooks⁴⁶ who also derived (117), we use the abbreviations

$$K_1 = \frac{1}{2} g_1 \beta H_{A1}; \quad K_2 = \frac{1}{2} g_2 \beta H_{A2}. \quad (118)$$

The expression (97) for the additive constant in the formula (95) for the eigenvalues now has E_0'' in place of E_0' . The expressions K_1 and K_2 can be regarded as effective anisotropy constants for the two sublattices. [An effective anisotropy energy of the form $K_i S_i \times \sin^2(\mathbf{S}_i, \mathbf{z}) = K_i S_i^{-1} (S_{ix}^2 + S_{iy}^2)$ is by (82), as modified in the foregoing, the same as $K_i \frac{1}{2} (P_i^2 + Q_i^2 - 1) = K_i n_i$, except for an additive constant, and so can be correlated with the H_{Ai} term in (115).]

The formulas for the frequencies of ferrimagnetic resonance absorption correspond to the frequencies of the completely symmetrical modes $\mathbf{k}=0$, and can hence be obtained from (117) by putting $\gamma_k=1$. The formulas thus obtained are more general than those first suggested by Kittel,⁴² as he assumed that the two sublattices had equal exchange and anisotropy fields. However, our calculation is still too special, as it assumes that the applied field is parallel to the anisotropy field. For arbitrary orientations, this situation is not true, and perhaps the most interesting part of the theory is the dependence on direction. The more general case is considered by Wangsness and others.⁴⁷ The resonance frequencies are there derived directly from the equations of motion, but doubtless the method of spin waves could be used instead. Wangsness⁴⁷ shows that the formula for the resonance frequencies in a ferrimagnetic material reduces to the conventional one for a ferromagnetic medium, provided the product of the molecular field coefficient and the net magnetization is large compared to the applied and anisotropy fields.

If the effects of the applied and anisotropy fields are relatively unimportant, the specific heat and the deviation in magnetic moment of a ferrimagnetic material should be proportional to $T^{\frac{3}{2}}$ at low temperatures, as in the case of a ferromagnetic material. This result perhaps at first sight seems rather surprising, because the presence of the radical sign presumably complicates the analysis. The point, however, is that at low temperatures only the modes with low characteristic frequencies are appreciably excited, and so only one branch, *viz.*, the branch $1k$ corresponding to the plus sign in (117), which has a vanishing frequency in the long wavelength limit when H , K_1 , and K_2 are zero, is of importance at low temperatures. If we take $K_1=K_2=0$, and if $1-\gamma_k^2 \ll 1$, the low frequencies of (117) are given by

$$\hbar \omega_{1k} = \frac{g_1 S_1 - g_2 S_2}{S_1 - S_2} \beta H + \frac{2J S_1 S_2}{S_1 - S_2} \sum_a a^2 k^2 \cos^2 \theta_{ka}. \quad (119)$$

⁴⁶ J. S. Kouvel and H. Brooks, Tech. Rept. 198, Cruft Laboratory, Harvard University, May 20, 1954 (unpublished).

⁴⁷ R. K. Wangsness, Phys. Rev. **91**, 1085 (1953); **93**, 68 (1954); **95**, 329 (1954); Am. J. Phys. **24**, 60 (1956).

This expression differs from the corresponding expression (22) for a ferromagnetic only in the constants of proportionality. So it is immediately seen that in place of (28) and (31) we now have

$$M_z = M_{z0} \left[1 - 0.1187 \left\{ \frac{kT(S_1 - S_2)}{2JS_1S_2} \right\}^{\frac{1}{2}} \right], \quad (120)$$

and

$$C_V = cNk \left(\frac{S_1 - S_2}{4JS_1S_2} \right)^{\frac{1}{2}} (kT)^{\frac{1}{2}}, \quad (121)$$

with c as in (31). The result (121) follows from (31) inasmuch as comparison of (119) and (22) shows that J is effectively modified by a factor $[S_1S_2/S(S_1 - S_2)]$ as compared with the ferromagnetic case. To obtain (120) from (28) we utilize in addition the fact that the deviation in magnetic moment per unit oscillator quantum number is modified by a factor $g^{-1}(g_1S_1 - g_2S_2)/(S_1 - S_2)$, as we see by comparing the coefficient of H in (119) and (22), and that M_{z0} has now $(g_1S_1 - g_2S_2)\beta$ instead of $g\beta$.

The results (120) and (121) were first obtained by Kouvel and Brooks⁴⁶ with a semiclassical analysis rather than the method of spin waves. The standard ferromagnetic formulas of Sec. IV correspond to the special case $S_1 = -S_2 = S$, as one might expect.

Specific heat of an antiferromagnetic.—An antiferromagnetic medium such as was studied in Sec. VII has $S_1 = S_2$. This case requires special treatment. The outstanding magnetization, of course, then vanishes in the absence of an applied field, and (119) cannot be used because of the zero denominator. Instead, the two branches $1k$ and $2k$ are now degenerate for $H=0$ and $K_1 = K_2 = 0$, and for vanishing applied field and anisotropy constants the low frequencies are now given by

$$\hbar\omega_k = [4J^2S^2z^2(1 - \gamma_k^2)]^{\frac{1}{2}} = 2JSka[z \sum_a \cos^2\theta_{k,a}]^{\frac{1}{2}}. \quad (122)$$

For a simple or body-centered cubic lattice [cf. discussion following Eq. (22)] this relation becomes

$$\hbar\omega_k = 2JSkl(2z)^{\frac{1}{2}}, \quad (123)$$

where z is the number of nearest neighbors. The formula for the intrinsic energy is hence, instead of (29),

$$\begin{aligned} U &= 2(L/2\pi)^3 4\pi \int_0^\infty \frac{[2JSkl(2z)^{\frac{1}{2}}]k^2 dk}{\exp(2JSkl(2z)^{\frac{1}{2}}/kT) - 1} \\ &= 2(L/l)^3 \frac{(kT)^4}{2\pi^2 [2JS(2z)^{\frac{1}{2}}]^3} \\ &\quad \times \int_0^\infty V^3(e^{-V} + e^{-2V} + \dots) dV. \end{aligned} \quad (124)$$

The initial factor 2 is a statistical weight to allow for the double degeneracy of the frequencies given by (96) when $H=0$. The specific heat $C_V = (dU/dT)$ corre-

sponding to (124) is

$$C_V = 2 \left(\frac{L}{l} \right)^3 \frac{1}{2\pi^2} \left(\frac{kT}{2JS(2z)^{\frac{1}{2}}} \right)^3 \zeta(4). \quad (125)$$

For a simple and body-centered cubic lattice the values of $(L/l)^3$ are N and $\frac{1}{2}N$, and of z are 6 and 8, respectively. The numerical value of (125) for the simple cubic case is

$$C_V = 13.7Nk(kT/12JS)^3, \quad (126)$$

a result also obtained by Kouvel and Brooks.⁴⁶

Comparison with experiment.—The predicted specific heats for ferrimagnetic and antiferromagnetic materials are thus strikingly different, being proportional to $T^{\frac{1}{2}}$ and T^3 , respectively. The $T^{\frac{1}{2}}$ dependence is confirmed in experiments made by Kouvel⁴⁸ on magnetite. His measurements on this ferrimagnetic nonconductor furnish perhaps the most direct experimental confirmation of the whole spin-wave theory. There are, of course, plenty of magnetic resonance experiments, but the resonance frequencies involve only the properties of very special symmetric states. It is easier to test the theory on specific heats than on the variation of magnetization with temperature, for the deviation of of the magnetization from its maximum value achieved at $T=0$ is small in the range of temperatures for which the spin-wave theory is applicable. Furthermore, measurements on ferrites are more definite than those on the usual ferromagnetic materials, inasmuch as the latter are conductors and so the results of the simple theory are complicated by the band structure, as we shall emphasize in Sec. XI. The exact theory which Kouvel gives for magnetite involves a somewhat different constant of proportionality in the $T^{\frac{1}{2}}$ law than that which we have developed, as this material has a spinel rather than simple cubic structure. Kouvel's interesting paper gives details and a discussion of the possibility of testing other results of the spin-wave theory experimentally. The T^3 formula for an antiferromagnetic involves the same form of temperature dependence as that for the specific heat arising from the lattice vibrations. Kouvel finds, however, that the magnetic contribution may involve a considerably larger proportionality constant than the ordinary Debye lattice term, and so should be capable of experimental detection. Corrections for the effect of anisotropy, however, spoil the rigor of the T^3 law.

X. HARMONIC OSCILLATOR MODEL FOR SPIN-RELAXATION PROCESSES

With the development of microwave spectroscopy, a great deal of interest has arisen in the relaxation processes by which the various spin states come into thermal equilibrium with each other and with the temperature of the lattice vibrations. Two characteristic relaxation times have to be distinguished, *viz.*,

⁴⁸ J. S. Kouvel, Phys. Rev. **102**, 1489 (1956).

the spin-spin and spin-lattice relaxation times. The former of these is usually the shorter, and arises from the dipolar or pseudodipolar interactions which spoil the constancy of the spatial components of magnetic moment. The latter arises from the modulation of either exchange or dipolar energies by the lattice vibrations. We do not go into the details of the theory of these relaxation processes; they are intricate and sometimes controversial as to whether the effects are large enough to be important. We do, however, want to outline how the various mechanisms can be formulated and intuitively visualized in terms of our harmonic oscillator model. The terms responsible for the relaxation are in each case anharmonic perturbations which induce transitions between the states of otherwise unperturbed harmonic oscillators.

First let us consider the case of spin-spin relaxation, as it is the simpler of the two. The anharmonic terms of the form (39) which arise from the $S_{ix}S_{jz}$ or $S_{iy}S_{jz}$ part of the dipolar or pseudodipolar potential, and which are nondiagonal in S_z' , couple together the different harmonic oscillators embodying the unperturbed spin waves. The problem of studying the processes of securing thermal equilibrium among the spin waves is thus mathematically similar to that of examining how the anharmonic terms perturb lattice vibrations and give rise to a finite thermal conductivity. The orders of magnitude which are calculated for the spin-spin relaxation times at high temperatures are reasonable, but ordinary models based on dipolar or pseudodipolar interaction lead to zero, or practically zero width of the magnetic resonance line at $T=0$, contrary to experiment. If all the magnetic atoms are similarly situated, the only interactions which contribute to line broadening are those which conserve energy and momentum of the spin waves. In a completely homogeneous material the broadening of a magnetic resonance line must hence arise from perturbations of the upper state, since the ground level has a lower energy than any other state. Thus the upper level can be broadened only by a third-order anharmonic term in which a symmetrical, or Kittel quantum (69) is de-excited and two spin waves of half the Kittel frequency and of mutually equal and opposite propagation vectors are excited. This process was suggested by Keffer and by Kittel and Abrahams.⁴⁹ In our opinion it is unlikely that it is frequent enough to give appreciable broadening at low temperatures. The same conclusion is also reached by Bloembergen and Wang,³³ and by Kasuya,⁵⁰ who investigates also still higher order anharmonic processes, and finds them ineffective near $T=0$.

The origin of the finite breadth of ferromagnetic resonance lines at low temperatures for a long time presented a perplexing problem. The mystery, however,

appears to have been removed recently in the case of the ferrites. In a material such as NiFe_2O_4 , the irregular distribution of the Ni^{++} and Fe^{+++} ions among the various octahedral sites furnishes the requisite inhomogeneity. Even so there was a difficulty. Until recently, it was thought that there were no other spin waves of frequency nearly equal to that of the symmetrical or Kittel wave $k=0$. If this is the case the perturbations caused by the irregularities are not potent enough to give an appreciable line width. However, Clogston, Suhl, Walker, and Anderson⁵¹ have now shown that the frequencies of the spin waves of low wave number are greatly distorted by the boundary conditions in specimens of finite size, and that as a result the degeneracy in the vicinity of $k=0$ which is prerequisite to line broadening can be obtained. The breadth should depend on how the body is cut, and so it does.

Previous to the paper of Clogston *et al.*, Galt, Yager, and Merritt⁵¹ proposed another mechanism for line breadth in those ferrites in which the octahedral sites contain both ferrous and ferric ions. Examples are Fe_3O_4 , or other ferrites containing excess iron as an impurity. The divalent and trivalent sites are then continuously redistributed by the process of electron transfer. A dynamic mechanism for damping and line broadening is thereby provided which is particularly effective when the transfer frequency is comparable with the signal frequency.

The broadening mechanisms for ferrites described in the two preceding paragraphs do not apply to ordinary metals such as nickel. A conceivable source of inhomogeneity and hence finite line width at $T=0$ for conducting metals may arise from an irregular distribution of atoms in different stages of ionization (d^8, d^9 , etc.) or possibly even from lattice imperfections. The requisite line breadth can be secured, however, only if the inhomogeneities do not fluctuate so rapidly as to be ineffective; electronic conduction may well average out the assignment of different valences to different lattice sites. A quite different mechanism for the breadth of ferromagnetic resonance lines has been proposed by Ament and Rado and was also intimated in an earlier analysis by MacDonald.⁵² These authors show that the skin effect damps the signal wave and broadens the resonance lines.

Further theoretical and experimental study of the spin-spin interactions as revealed by the width of the resonance lines is desirable. The experiments of Gerritsen⁵³ on antiferromagnetic resonance indicate that in antiferromagnetics the line width tends to zero at $T=0$.

Garnets differ from ferrites in that there is no possibility in the garnets of redistributions of ions of

⁴⁹ F. Keffer, Phys. Rev. **88**, 686 (1952); C. Kittel and E. Abrahams, Revs. Modern Phys. **25**, 233 (1953).

⁵⁰ T. Kasuya, Progr. Theoret. Phys. **12**, 802 (1954).

⁵¹ Galt, Yager, and Merritt, Phys. Rev. **93**, 1119 (1954); A. M. Clogston, Bell System Tech. J. **34**, 739 (1955).

⁵² W. S. Ament and G. T. Rado, Phys. Rev. **97**, 1558 (1955); Proc. Phys. Soc. (London) **A64**, 968 (1951).

⁵³ Gerritsen, Garber, and Drewes, Physica **22**, 213 (1956).

different valences among the different lattice sites, and it is indeed found that they have exceedingly narrow resonance lines.

Let us now turn to spin-lattice coupling. The spin-lattice relaxation arises from the modulation of the spin interaction energies i.e., the spin waves by the crystalline vibrations. We can expand the exchange integrals J_{ij} and the dipolar constants D_{ij} which figure in our various equations such as (1), (2), and (3), as power series in the normal coordinates representing the lattice distortions, and retain only linear terms in this development. As the original Hamiltonian before such an expansion is of the second order in the spin-wave oscillator coordinates and momenta, we thus obtain a perturbing Hamiltonian of the form

$$\sum_{ijk}(a_{ijk}Q_iQ_jq_k+b_{ijk}P_iP_jq_k), \quad (127)$$

where the q_k are phonon and the Q_i and P_i spin-wave oscillator coordinates. Since both the spin waves and the phonons can be interpreted as harmonic oscillators, we have again the situation of a third-order anharmonic perturbative coupling of a system of harmonic oscillators. The difference as compared with spin-spin interaction is that in the spin-lattice interaction one of the quanta is a phonon and two are spin waves, whereas in the spin-spin interaction all three are spin waves. (Besides the third order, there may be even higher order processes, but we assume for simplicity that the third-order effect is the most important one.) Terms of the structure (127) can arise from purely isotropic exchange coupling, whereas in spin-spin interaction the significant perturbations are of necessity those which are nondiagonal in the magnetization.

In an important paper,⁵⁴ Akhieser examined the spin-spin and spin-lattice relaxation effects in considerable detail. His calculations have, however, been criticized by Polder,⁵¹ who claims that when properly revised they do not yield sufficiently short relaxation times to agree with experiment. Akhieser used purely classical dipolar interaction, and Kittel and Abrahams^{49,55} point out that larger perturbations are possible because of pseudodipolar coupling arising from spin-orbit interaction. They had the ingenious idea of taking the constants from the empirical magneto-elastic constants. In this fashion they claim to obtain sufficient spin-lattice interaction. The way in which Akhieser replaced sums by integrals probably tended to underestimate the effect of the modulation of short-range exchange integrals, which depend exponentially on distance.

XI. QUESTIONS OF VALIDITY OF SPIN-WAVE THEORY OUTSIDE THE HEITLER-LONDON MODEL

So far we have presented spin waves on the basis of the Heitler-London, or Heisenberg model, wherein

the spins are localized at regularly spaced lattice sites. The question arises whether these waves can also be used in more general models of a solid, such as for instance the conduction band model. Herring and Kittel⁵⁶ have shown that from a purely mathematical standpoint the periodic transformation (17) associated with spin waves can be used to help diagonalize the energy in any solid with regularly spaced atoms, even though the electrons wander from one atom to another. However, in our opinion spin waves must be used with caution in an actual conductor. One cannot necessarily say, for instance, that in the conduction band model the energy can be regarded as the sum of the Sommerfeld translational energy, and a spin-wave energy obeying the $T^{\frac{3}{2}}$ law for specific heat. The essence of the usual spin-wave theory is that in the absence of an external field energy quanta associated with spin waves of low propagation wave number k vary quadratically with k [cf. Eq. (22)]. Expressed differently, the lowest energy levels among the states of spin $S=NS-1$ are contiguous to the energy of the state of maximum spin $S'=NS$. In an extreme conduction band model, the exchange integral is practically the same between all states. This can be seen from a paper of Slater,⁵⁷ or more intuitively from the fact that in an ideal conductor all electrons travel uncorrelated through the lattice, so that no pairs of states stand in a preferred or neighborly relation. Under these circumstances the energy is determined only by the total spin, for if J_{ij} is independent of i and j , the spin-coupling energy is

$$\begin{aligned} -2\sum_{j>i}J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j &= -2J\sum_{j>i}\mathbf{S}_i\cdot\mathbf{S}_j \\ &= -J[(\sum_i\mathbf{S}_i)^2-\sum_i(\mathbf{S}_i)^2] \\ &= -J[S'(S'+1)-NS(S+1)]. \end{aligned} \quad (128)$$

There is then a high degree of degeneracy, and not a spreading of the energy values for given $S'=NS-1$, such as one associates with spin waves. The quantum number S' , which is the quantum number quantizing the resultant magnitude of the spin of the entire crystal, should not be confused with the quantum number S'_z describing a spatial component of this total spin. The usual spin-wave calculation does not mention this spin quantum number S' , but as a matter of fact, of the N eigenvalues for the rigorously soluble case $S'_z=NS-1$, the symmetric solution $\mathbf{k}=0$ belongs to $S'=NS$, and the remaining $N-1$ states to $S'=NS-1$. In the situation corresponding to Eq. (128), all these remaining states have the same energy. In terms of the spin-wave calculation, this state of affairs arises because the spin secular determinant has identical entries in all places, instead of being essentially a border determinant, as is the case when only adjacent atoms are appreciably coupled. The proof that the spin-wave solution reduces to (128), as of course it

⁵⁴ A. Akhieser, J. Phys. (U. S. S. R.) 10, 217 (1946).

⁵⁵ E. Abrahams and C. Kittel, Phys. Rev. 88, 1200 (1952).

⁵⁶ C. Herring and C. Kittel, Phys. Rev. 81, 869 (1951).

⁵⁷ J. C. Slater, Phys. Rev. 49, 537 (1936).

must, is as follows. When all the matrix elements are identical, the spin-wave solution corresponding to (20) is, apart from an additive constant,

$$E = -2S \sum_{ij} J_{ij} \exp(i\mathbf{k} \cdot \mathbf{R}_{ij}) = -2JNS \sum_j \exp(i\mathbf{k} \cdot \mathbf{R}_{ij}).$$

In virtue of the property (49) of the lattice waves, this formula for the energy levels becomes

$$\begin{cases} E = -2JNS & \text{for } k=0; \\ E = 0 & \text{for } k \neq 0. \end{cases} \quad (129)$$

Thus instead of being a virtual continuum of states, there are only two different energy values. The difference of the two roots (129) agrees exactly with the difference of the general expression (128) evaluated for $S' = NS$ and $S' = NS - 1$.

We can thus say that, so to speak, the spin-wave properties are essentially expurgated when all the entries in the secular determinant are similar. That this state of affairs should occur is not surprising. The reason that in the ordinary spin-wave theory for the Heitler-London model the energy is practically the same for $S' = NS$ and the deepest states belonging to $S' = NS - 1$ is that in the latter the average orientation of the spins varies very slowly with position, the total variation being enough to make the resultant spin be $S' = NS - 1$ rather than $S' = NS$. Thus, two atoms which are adjacent have their spins practically parallel. However, the essence of the band model is that the electron in a given state is distributed equally over all atomic states. Hence it is impossible to localize the distribution of spin orientations and bend the electron spin vector around slowly from place to place. One can say that electron correlation (in the language of solid state physicists) is a prerequisite to meaningful spin waves.

At this stage, the reader may feel that all our preceding discussion of spin waves is wasted in conductors, since the literal band picture leads to the energy structure (129), in which all vestiges of spin waves are suppressed. This, however, is too pessimistic a view. The result (129) is obtained only if one disregards all matrix elements of the exchange energy which are nondiagonal in the orbital translational quantum numbers k_{x1} , k_{y1} , k_{z1} of the individual electrons. Actually the exchange energy of any pair of electrons 1, 2, though diagonal in $\mathbf{k}_1 + \mathbf{k}_2$ (modulus umklapp processes), because of the conservation of momentum, need not be so in the quantum numbers of the individual electrons. Some of these elements need not be very much off-diagonal in energy. One can blend together states of slightly different translational energy so as to construct a wave packet whose spin is zero, and whose translational energy is only slightly different from that of a pure translational state. Suppose, for instance, one had a very large piece of conductor, and assumed that in one half of it the spins all pointed down, and in the other half up. Then the resultant spin would

be zero, and the exchange energy not too different from the state in which the spins were all parallel. If the material is ferromagnetic, i.e., favors parallel alignment of the spins, there is a highly unfavorable exchange energy connecting the spins on the opposite sides of the boundary separating the two halves. It would, of course, be better to make the transition from the north to the south pointing spins a gradual rather than sudden affair, as then adjacent spins are nearly parallel. In fact, one can develop a continuum theory⁵⁶ on this basis, on which the exchange energy can be shown to be proportional to $\nabla^2 M$, where M is the magnetization vector. If the material is anisotropic, the transition in the spins should not be too gradual, as then the spin is of necessity unfavorably oriented in certain portions of the turn-over region, and these portions should not be allowed to get too wide. Such considerations, however, would take us into study of the thickness of the Bloch wall, domain theory, etc., subjects into which we do not intend to enter. If the resultant spin for the entire body is only slightly less than its maximum permitted value, then one can think of the local average direction of magnetization as spiraling around the direction of total magnetization. This is essentially a statistical spin-wave model. A sort of generalized spin-wave theory, founded on the basis of average properties in a conductor, has been developed by Herring and Kittel.⁵⁵ The details are rather involved, and the legitimacy of some of the approximations not very transparent.

The gist of the preceding discussion is that the papers of Herring and Kittel make it plausible that the results of the spin-wave theory apply at least qualitatively to conductors. This is particularly true if one is in the ferromagnetic state, as then the state of maximum spin is one in which fluctuations in the charge per atom are kept to a minimum because of the exclusion principle. In fact, if there is only one permitted orbital state per atom and one conduction electron per atom, no fluctuations at all in the charge per atom is permitted if the spins are all parallel. Obviously the degeneracy of the d sites, a complication not considered in simple spin-wave theory, is an important and complicating factor. In our opinion, in the statistical spin-wave construction found in the Kittel-Herring paper, there is a suppression of states of higher polarity and a degree of localization established which can in many ways be regarded as a concealed reversion to essentially the Heitler-London nonpolar model.

The generalized analysis of Kittel and Herring indicates that there may be elements of truth in the spin-wave theory even in conductors, and so casts considerable doubt on the treatment of the magnetism of metals by the "collective electron ferromagnetism" model of Stoner.⁵⁸ The essential approximation of this

⁵⁶ E. C. Stoner, Proc. Roy. Soc. (London) **A165**, 372 (1938); **A169**, 339 (1939); E. P. Wohlfarth, Revs. Modern Phys. **25**, 211 (1953).

model that exchange interaction can be represented by means of a molecular field is more or less tantamount to the result (129) based on uncorrelated electron migrations. In this connection one can recall the result in Heisenberg's first paper⁴ on magnetism, that molecular field theory is equivalent to the assumption that all states with the same spin have the same energy. The conclusions of Stoner concerning the variation with temperature of the exchange specific heat are therefore suspect. He found that the exchange specific heat should be proportional to T , and that the deviation of the saturation intensity of magnetization from its maximum value at $T=0$ should be proportional to T^2 , in contrast to the T^3 factor obtained in both cases by spin-wave theory. As a matter of fact there is, incidentally, little, if any evidence, in favor of the T and T^2 laws. (In the case of the specific heat translational effects mask the exchange contributions.)

The reader should not, however, form the impression that the results of the spin-wave theory hold at least qualitatively under any metallic situation, for the generality is undoubtedly not that great. If, for instance, the number of electrons, or alternatively of "holes" relative to closed configurations is small compared with the number of atomic sites, the fluctuation effects presumably become rather tremendous, and the concepts of spin correlation and density begin to lose meaning. Just where this difficulty arises quantitatively is at present anything but clear. It is true that very general considerations suggest wide validity for perhaps the most significant result of spin-wave theory,⁵⁹ namely that the energy of long spin waves be proportional to the square of the frequency or propagation wave number k . If the origin is taken at the state of

completely parallel alignment the interaction exchange energy is proportional to $J(1-\cos\Delta\varphi)$ where $\Delta\varphi$ is the angle between the directions of the mean magnetization at the sites 1 and 2 [cf. Eq. (1)]. The essential point is that if k is small, $\Delta\varphi$ is proportional to k and so $1-\cos\Delta\varphi$ is proportional to k^2 . However, if the electron spins are irregularly distributed, both in position or time, the concept of a well-defined wavelength may lose meaning. The difficulties encountered in trying to generalize spin-wave theory to conductors or other arrays of irregularly spaced electrons may be likened to those which one would experience in trying to extend the usual Born-Karman theory of lattice vibrations to the case where the atoms are of various varieties irregularly spaced and continually exchanging places. Just how long the Debye T^3 law for lattice specific heats retains its validity under such circumstances is something of a moot question. At least the authors are not aware of an appropriate analysis. So also it is with the T^3 laws characteristic of spin-wave theory when the spin distribution becomes too irregular. Any mathematical formalism for such more complicated situations is clearly quite another matter than the conventional spin-wave theory for uniformly spaced spins, and so it is well to end the present paper here. We may, however, terminate by remarking that the ideal materials for testing the standard theory are the ferrites. Their nonconducting properties make them indeed a boon to the theoretical physicist as well as to the engineer. Garnets are even better.

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⁵⁹ Compare C. Kittel, Varenna Lectures, *Nuovo cimento* (to be published).