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Theory of Spin-Wave Interactions in Ferro- and Antiferromagnetism*

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The spin-wave theory in an ideal Heisenberg model of a ferromagnet is studied using Holstein and Primakoff's method including the spin-wave interactions. Several earlier published results of the correction to the spontaneous magnetization produced by spin-wave interactions were in disagreement with each other, and they were not in agreement with Dyson's result which is regarded as rigorous at low temperatures. Our result is in agreement with Dyson's to the order which we have considered.

Our method can be applied to antiferromagnetism easily. We have obtained the correction arising from interactions between spin waves. The correction term is quite small. This means that the simple theory neglecting the spin-wave interactions is sufficient for practical purposes.

I. INTRODUCTION

BLOCH[1] first presented a spin-wave theory in the Heisenberg model of ferromagnetism. He predicted the temperature dependency of the magnetization at low temperatures (Bloch's $T^{\frac{3}{2}}$ law). In his theory he assumes that the number of spin waves is so small that the interaction between two or more spin waves may be neglected. The same result was obtained by Holstein and Primakoff[2] (H-P) using creation and annihilation operators of a spin wave under the same assumption as Bloch's.

Dyson[3] invented a general theory of spin-wave interactions. In his theory he defines two kinds of interactions: One is the kinematical interaction which arises from the fact that more than $2S+1$ units of reversed spin (S is the magnitude of atomic spin in units of $\hbar$) cannot be attached to the same atom simultaneously. The other is the dynamical interaction which represents the nondiagonal part of the Hamiltonian in his basic set of states. The merit of his treatment is that the kinematical interaction can be neglected in an expansion in powers of $\kappa T/J$ (κ, T and J are Boltzmann's constant, the temperature and the exchange integral, respectively) and that the dynamical interaction is very weak at low temperatures. Dyson also criticizes H-P spin-wave theory in that although the kinematical interaction does not appear, the dynamical interaction is so strong in their treatment that one cannot get rid of mathematical difficulties.

Prior to Dyson's paper several authors[4-7] obtained correction terms in the expression of the spontaneous magnetization of ferromagnetism at low temperatures. Among these authors Schafroth[5] and Heber[6] followed the ideas of H-P. They were neither in agreement with each other nor with Dyson. We believe that the origins of their incorrect results are not in H-P's method itself, but due to their poor approximations.

Although Dyson's paper is rigorous, it is not so easy to understand. We will show in Sec. II that a careful treatment of H-P's method gives the same results (to first order in $1/S$) as Dyson's, and is very simple. Mannari and Terasaki[8] and Morita[9] have obtained essentially the same conclusions independently of the author.

H-P's method has been applied to antiferromagnetism by Kubo.[10] He has obtained the expressions of many thermodynamical quantities and discussed the divergence of some variables in the absence of anisotropy energy. Our method can be applied to antiferromagnetism easily, and the results will be shown in Sec. III.

II. FERROMAGNETISM

The nearest neighbor exchange interaction model of a ferromagnet is described by the following Hamiltonian,

$$\mathcal{H}=-2J\sum_{\langle j,l\rangle}\mathbf{S}_j\cdot\mathbf{S}_l+Hg\mu\sum_{j=1}^{N}S_j^z. \tag{1}$$

Here $\mathbf{S}_j$ is the spin operator at the jth atom, N the total number of atoms, g the Lande g-factor, μ the Bohr magneton, the summation $\sum_{\langle j,l\rangle}$ is taken over all nearest neighbor pairs, and the external magnetic field H is directed along the z axis. Following H-P, we express the spin operators in the forms,

$$\begin{aligned} S_j^+&=S_j^x+iS_j^y=(2S)^{\frac{1}{2}}f_j(S)a_j,\\ S_j^-&=S_j^x-iS_j^y=(2S)^{\frac{1}{2}}a_j^*f_j(S),\\ S_j^z&=S-n_j, \end{aligned} \tag{2}$$

where

$$f_j(S)=(1-n_j/2S)^{\frac{1}{2}}, \tag{3}$$

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and the operator $a_j^*a_j=n_j$ is called the spin deviation operator, and a_j^* and a_j are to be regarded as the creation and annihilation operator of the spin deviation, and they satisfy the commutation rule

$$a_ja_l^*-a_j^*a_l=\delta_{jl}. \tag{4}$$

Using these operators the Hamiltonian (1) can be written as follows,

$$\mathcal{H}=-2J\sum_{\langle j,l\rangle}[2Sa_j^*f_j(S)f_l(S)a_l-2Sa_j^*a_j+a_j^*a_ja_l^*a_l]-Hg\mu\sum_j a_j^*a_j, \tag{5}$$

where we have dropped the constant terms. We would like to make the following remarks about the operator $f_j(S)$. There are several definitions for $f_j(S)$. One is the binomial expansion in the Hilbert space of infinite dimensions such as

$$f_j(S)=1-\frac{n_j}{4S}-\frac{1}{32}\frac{n_j^2}{S^2}\cdots. \tag{6}$$

A second is the following expansion in the same space as (6),

$$f_j(S)=\sum_{m=0}^{\infty}c_m(S)a_j^{*m}a_j^{m}, \tag{7}$$

where the $c_m(S)$ have to be determined from a recurrence relation. Expressions (6) and (7) are equivalent. A third one is defined in the subspace of $(2S+1)$ dimensions for $0\leq n_j\leq 2S$, as follows,

$$f_j(S)=\sum_{m=0}^{2s}d_m(S)n_j^{m}, \tag{8}$$

where the $d_m(S)$ can be determined individually. As n_j is the spin deviation, it loses physical meaning if it is greater than $2S+1$. Considering this situation, Eq. (8) can be best used for rigorous treatments of the Hamiltonian (5). However rigorous treatments are so difficult that Eq. (8) is not very convenient. On the contrary, the limiting of $f_j(S)$ to the subspace as in Eq. (8) leads to the more tedious formalism. As H-P mentioned already and Dyson showed definitely the states $n_j\geq 2S+1$ give a negligibly small contribution at low temperatures, so that we may neglect the kinematical interactions. A glance at Eq. (6) shows that it is an expansion of the powers of not only n_j but also $1/S$, which is very important, as will be shown later. Thus we will use Eq. (6) in this paper.

We introduce the Fourier transforms of a_j and a_j^*,

$$a_k=(1/N^{\frac{1}{2}})\textstyle\sum_j e^{-ikj}a_j,\quad a_k^*=(1/N^{\frac{1}{2}})\sum_j e^{ikj}a_j^*, \tag{9}$$

then we obtain the transformed Hamiltonian as

$$\mathcal{H}=\mathcal{H}^0+\mathcal{H}', \tag{10}$$

$$\mathcal{H}^0=\textstyle\sum_k A_ka_k^*a_k, \tag{11}$$

$$A_k=2SJz(1-\gamma_k)-Hg\mu, \tag{12}$$

$$\begin{aligned}\mathcal{H}'=&\sum_{k_1,\cdots,k_4}\delta(k_1+k_2-k_3-k_4)\\&\times\left[\frac{Jz}{2N}(\gamma_1+\gamma_4-2\gamma_{2-4})+\frac{Jz}{16SN}(\gamma_1+\gamma_4)\right]\\&\times a_1^*a_2^*a_3a_4+\frac{Jz}{16SN^2}\sum_{k_1,\cdots,k_6}\\&\times\delta(k_1+k_2+k_3-k_4-k_5-k_6)\\&\qquad\times(\gamma_1+\gamma_6-2\gamma_{3-5-6})a_1^*a_2^*a_3^*a_4a_5a_6+\cdots,\end{aligned} \tag{13}$$

where we have defined

$$\gamma_k=\textstyle\sum_\rho e^{ik\rho}/z, \tag{14}$$

with ρ denoting the vectors to the nearest neighbor atoms, and z the number of them. We have used the notation a_1, γ_1 etc. instead of a_{k_1}, γ_{k_1} etc., and later we will use n_1, A_1 etc. instead of n_{k_1}, A_{k_1} etc.

We will use the following basic set of states,

$$\Psi\{n_k\}=\prod_k\frac{1}{(n_k!)^{\frac{1}{2}}}(a_k^*)^{n_k}|0\rangle,\quad n_k=0,1,2,\cdots; \tag{15}$$

here $|0\rangle$ is the state of no spin waves. The basic states (15) are normalized and orthogonal to each other. If we operate with the Hamiltonian (10) on the states (15), the result is

$$\begin{aligned}&\mathcal{H}\Psi\{n_k\}\\&=\left\{\sum_k A_kn_k+\sum_{k_1,k_2}\left[\frac{Jz}{N}(\gamma_1+\gamma_2-1-\gamma_{1-2})+\frac{Jz}{8SN}(\gamma_1+\gamma_2)\right]n_1n_2+\sum_{k_1,k_2,k_3}\right.\\&\quad\left.\times\frac{Jz}{4SN^2}(\gamma_1-\gamma_{3-2-1})n_1n_2n_3+\cdots\right\}\\&\quad\times\Psi\{n_k\}+\sum_{k_1,\cdots,k_4}{}'\,\delta(k_1+k_2-k_3-k_4)\\&\quad\times\left[\frac{Jz}{2N}(\gamma_1+\gamma_4-2\gamma_{2-4})+\frac{Jz}{16SN}(\gamma_1+\gamma_4)\right]\\&\qquad\qquad\times\Psi\{n_k'\}+\cdots,\end{aligned} \tag{16}$$

where $\sum'$ means we exclude the terms of $k_1=k_3$, $k_2=k_4$ and $k_1=k_4$, $k_2=k_3$, and $\Psi\{n_k'\}$ etc. are different states from $\Psi\{n_k\}$, i.e., they express the dynamical interactions. These dynamical interactions are not so weak as those in Dyson's treatment, and he therefore did not treat Eq. (16). However, the diagonal terms of Eq. (16) are also not so simple as those in Dyson's Eq. (26) in his first paper, and fortunately many of them are canceled with nondiagonal terms in the following treatment, and we will get the same result as Dyson gets.

In Eq. (10) $\mathcal{H}^0$ is the Hamiltonian of free spin waves, and $\mathcal{H}'$ represents the interactions between spin waves. It is by no means clear that $\mathcal{H}'$ is smaller than $\mathcal{H}^0$ at this stage. However, if we regard Eq. (10) as an expansion of powers of $1/S$, we can surely treat $\mathcal{H}'$ as a perturbation. In the following we will calculate the grand partition function of the system defined from

$$Z=\text{Trace}\exp(-\beta\mathcal{H}),\quad \beta=1/(\kappa T),\tag{17}$$

where we have taken the chemical potential as zero. As is well known,[11] Eq. (17) can be developed into the following form,

$$\begin{aligned}Z=\text{Trace}\biggl\{&\exp(-\beta\mathcal{H}^0)\biggl[1-\int_0^\beta\exp(s\mathcal{H}^0)\mathcal{H}'\\&\times\exp(-s\mathcal{H}^0)ds+\int_0^\beta\exp(s\mathcal{H}^0)\mathcal{H}'\exp(-s\mathcal{H}^0)\int_0^s\\&\times\exp(t\mathcal{H}^0)\mathcal{H}'\exp(-t\mathcal{H}^0)dtds+\cdots\biggr]\biggr\}.\end{aligned}\tag{18}$$

In this expansion we will calculate the terms to order $1/S$. The first term in the parentheses gives, of course, the ground partition function of free spin waves as follows,

$$Z_{\text{I}}=\sum_{n_k=0}^{\infty}\exp(-\beta\sum_k A_kn_k).\tag{19}$$

In the second term only the diagonal terms of $\mathcal{H}'$ do not vanish, and the result is

$$\begin{aligned}Z_{\text{II}}=-\beta\sum_{n_k=0}^{\infty}&\exp(-\beta\sum_k A_kn_k)\\&\times\biggl[-\frac{Jz}{N}\sum_{k_1,k_2}(1+\gamma_{1-2}-\gamma_1-\gamma_2)n_1n_2\\&+\frac{Jz}{8SN}\sum_{k_1,k_2}(\gamma_1+\gamma_2)n_1n_2\\&+\frac{Jz}{4SN^2}\sum_{k_1,k_2,k_3}(\gamma_1-\gamma_{3-2-1})n_1n_2n_3+O(1/S^2)\biggr].\end{aligned}\tag{20}$$

In the third term the nondiagonal terms contribute to the result,

$$\begin{aligned}Z_{\text{III}}=\sum_{n_k=0}^{\infty}&\exp(-\beta\sum_k A_kn_k)\biggl[\frac{Jz}{8SN^2}\sum_{k_1,k_2,k_3}\\&\times\frac{|\gamma_1+\gamma_2+\gamma_3+\gamma_{1+2-3}-2\gamma_{1-3}-2\gamma_{2-3}|^2}{\gamma_1+\gamma_2-\gamma_3-\gamma_{1+2-3}}\\&\times(n_1n_2+2n_1n_2n_3)+O(1/S^2)\biggr].\end{aligned}\tag{21}$$

Further terms give nothing to order $1/S$. In the following calculations we will confine our consideration to the simple cubic lattice. In Eqs. (20) and (21) the terms in n_1n_2 express two spin-wave interactions, and combine to give the following term after a simple calculation,

$$\begin{aligned}\sum_{n_k=0}^{\infty}&\exp(-\beta\sum_k A_kn_k)\frac{\beta J}{2N}\sum_{k_1,k_2}\\&\times(k_{1x}^2k_{2x}^2+k_{1y}^2k_{2y}^2+k_{1z}^2k_{2z}^2)\\&\times\biggl[1+\frac{1}{S}\biggl(\frac{1}{6}+\frac{7}{216}+\cdots\biggr)\biggr]n_1n_2.\end{aligned}\tag{22}$$

The term in $n_1n_2n_3$ in Eq. (21) corresponds to three spin-wave interactions; it will give higher order contributions and will not be analyzed further. Thus the grand partition function can be written from Eqs. (19) to (22) as follows,

$$\begin{aligned}Z=\prod_k(1-e^{-\beta A_k})^{-1}&\biggl[1+\frac{\beta J}{2N}\sum_{k_1,k_2}\\&\times(k_{1x}^2k_{2x}^2+k_{1y}^2k_{2y}^2+k_{1z}^2k_{2z}^2)\\&\times\biggl(1+\frac{0.2}{S}\biggr)\times(e^{\beta A_1}-1)^{-1}(e^{\beta A_2}-1)^{-1}\biggr].\end{aligned}\tag{23}$$

By the standard method the spontaneous magnetization M is given by

$$\begin{aligned}\frac{M}{Ng\mu}=S-\zeta(\tfrac{3}{2})\biggl(\frac{\kappa T}{8\pi JS}\biggr)^{3/2}&-\frac{3\pi}{4}\zeta(\tfrac{5}{2})\biggl(\frac{\kappa T}{8\pi JS}\biggr)^{5/2}\\&-\frac{33\pi^2}{32}\zeta(\tfrac{7}{2})\biggl(\frac{\kappa T}{8\pi JS}\biggr)^{7/2}-\frac{3\pi}{2S}\biggl(1+\frac{0.2}{S}\biggr)\\&\times\zeta(\tfrac{3}{2})\zeta(\tfrac{5}{2})\biggl(\frac{\kappa T}{8\pi JS}\biggr)^{4}\cdots,\end{aligned}\tag{24}$$

where $\zeta(n)$ is the Riemann zeta function

$$\zeta(\tfrac{3}{2})=2.612,\quad \zeta(\tfrac{5}{2})=1.341,\quad \zeta(\tfrac{7}{2})=1.127.\tag{25}$$

In Eq. (24) the term in $T^{3/2}$ corresponds to the $T^{3/2}$ law

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[11] For example, see M. L. Goldberger and E. N. Adams, II, J. Chem. Phys. **20**, 240 (1952) and R. Serber, Phys. Rev. **43**, 1011 (1933).

of Bloch, as it should, and the terms in $T^{5/2}$ and $T^{7/2}$ are due to the corrections of the long-wavelength approximation. The term in T^4 corresponds to Dyson's correction term, although our numerical factor contains $(1+0.2/S)$, whereas Dyson's contains $(1+0.3/S)$. The difference may come from numerical approximations in Eq. (22) in our paper and in Eq. (92) in Dyson's first paper. (Note that J used in the present paper is just half Dyson's J.)

Thus we have obtained an essentially same result as Dyson's, using the spin-wave formalism of H-P. We will further show the equivalence of the two methods in another way. In Eq. (10) we rearrange the terms as follows:

$$\mathcal{H}=\mathcal{H}_D+\mathcal{H}_R, \tag{26}$$

$$\mathcal{H}_D=-Hg\mu\sum_j a_j^*a_j+2SJ\sum_{\langle j,l\rangle}(a_j^*-a_l^*)(a_j-a_l)+J\sum_{\langle j,l\rangle}a_j^*a_l^*(a_j-a_l)^2, \tag{27}$$

$$\begin{aligned}\mathcal{H}_R=J\sum_{\langle j,l\rangle}&(a_j^*a_j^*a_ja_l-a_j^*a_l^*a_ja_j)\\&+\frac{J}{8S}\sum_{\langle j,l\rangle}(a_j^*a_j^*a_ja_l+a_j^*a_l^*a_la_l)\\&+\frac{J}{8S}\sum_{\langle j,l\rangle}(a_j^*a_j^*a_j^*a_ja_ja_l-2a_j^*a_j^*a_l^*a_ja_la_l\\&\qquad+a_j^*a_l^*a_l^*a_la_la_l)+\cdots.\end{aligned} \tag{28}$$

$\mathcal{H}_D$ is just the same as Dyson's Hamiltonian [Eq. (57) in his first paper]. After a Fourier transform the first term in Eq. (28) gives

$$\frac{zJ}{2N}\sum_{k_1,\cdots,k_4}\delta(k_1+k_2-k_3-k_4)a_1^*a_2^*a_3a_4(\gamma_4-\gamma_2). \tag{29}$$

The diagonal terms of Eq. (29) vanish, and the contribution from nondiagonal terms is given by

$$-\frac{zJ}{4SN}\sum_{k_1,k_2}\gamma_1n_1n_2-\frac{zJ}{4SN^2}\sum_{k_1,k_2,k_3}(\gamma_3-\gamma_{2+3-1})n_1n_2n_3. \tag{30}$$

However, the diagonal parts of the second and third terms in Eq. (28) cancel the first and second terms in Eq. (30), respectively. Thus $\mathcal{H}_R$ (Remainder of the Hamiltonian) gives nothing to order $1/S$. This means $\mathcal{H}$ is equivalent to $\mathcal{H}_D$ at least to order $1/S$.

III. ANTIFERROMAGNETISM

As is well known the exchange integral in antiferromagnets is negative so that an antiparallel spin coupling between nearest neighbor atoms is more stable than a parallel one. However, the ground state in Heisenberg's model of the antiferromagnetic case is very complicated and is known only approximately, while the ground state in the ferromagnetic case is very simple and is a starting point of a spin-wave theory in ferromagnetism. In spite of this the successes of spin-wave theories of antiferromagnetism by Anderson[12] and Kubo[10] make us feel that they contain a correct approach to attack the problem. As was discussed by Kubo in detail there is a divergence of the fluctuation of magnetization in spin-wave theory of antiferromagnetics. This divergence can be avoided by introducing the anisotropy energy. In fact we cannot doubt the importance of the role of the anisotropy energy in antiferromagnetism. However, this divergence of the fluctuation occurs in the spin-wave theory of ferromagnetism, too. On the other hand our method used in the last section is essentially the same as Dyson's in ferromagnetism. We will apply it to an ideal antiferromagnet with no anisotropy energy, which may correspond to Dyson's method if it were done for antiferromagnetism.

Let us confine our considerations to a NaCl type and a CsCl type crystal, and divide the lattice points into two sublattices, denoted by l and m, respectively. The Hamiltonian is assumed to be

$$\mathcal{H}=2|J|\sum_{\langle l,m\rangle}(\mathbf{S}_l\cdot\mathbf{S}_m)+h_1\sum_l S_l^z+h_2\sum_m S_m^z, \tag{31}$$

with different effective magnetic field on l and m sublattices. The reason for using different magnetic field parameters is that by differentiation with respect to a single parameter we may select out the magnetization of a sublattice. According to Kubo we have to introduce two different definitions of the spin-deviation operators. These are

$$S_l^+=(2S)^{\frac{1}{2}}f_l(S)a_l,\quad S_l^-=(2S)^{\frac{1}{2}}a_l^*f_l(S),\quad S_l^z=S-a_l^*a_l, \tag{32}$$

for an "up" spin on the l sublattice, and

$$S_m^+=(2S)^{\frac{1}{2}}b_m^*f_m(S),\quad S_m^-=(2S)^{\frac{1}{2}}f_m(S)b_m,\quad S_m^z=-S+b_m^*b_m, \tag{33}$$

for a "down" spin on the m sublattice. The operators b and b^* are defined in the same way as a and a^*, and satisfy the equations,

$$b_m^*b_m=n_m',\quad b_mb_m^{*\prime}-b_m^*b_m'=\delta_{mm'}. \tag{34}$$

We introduce the Fourier transforms of a_l, a_l^* and b_m, b_m^*:

$$\begin{aligned}&a_k=(2/N)^{\frac{1}{2}}\textstyle\sum_l e^{ikl}a_l,\qquad a_k^*=(2/N)^{\frac{1}{2}}\sum_l e^{-ikl}a_l^*,\\&b_k=(2/N)^{\frac{1}{2}}\textstyle\sum_m e^{-ikm}b_m,\ b_k^*=(2/N)^{\frac{1}{2}}\sum_m e^{ikm}b_m^*,\end{aligned} \tag{35}$$

where N is the total number of atoms. Furthermore, we use the new operators α_k, α_k^* and β_k, β_k^* defined by

$$\begin{aligned}a_k&=\alpha_k\cosh\theta_k-\beta_k^*\sinh\theta_k,\\b_k&=-\alpha_k^*\sinh\theta_k+\beta_k\cosh\theta_k,\\a_k^*&=\alpha_k^*\cosh\theta_k-\beta_k\sinh\theta_k,\\b_k^*&=-\alpha_k\sinh\theta_k+\beta_k^*\cosh\theta_k,\end{aligned} \tag{36}$$

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[12] P. W. Anderson, Phys. Rev. **86**, 694 (1952).

where

$$\tanh 2\theta_k=\gamma\kappa/(1-\tfrac{1}{2}h_1'+\tfrac{1}{2}h_2'), \tag{37}$$

$$h_1'=h_1/2z|J|S, \quad h_2'=h_2/2z|J|S. \tag{38}$$

Then the Hamiltonian (31) can be written as

$$\mathcal{H}=E_h+\sum_k (A_k^-n_k+A_k^+n_k') + \sum_{k_1k_2} [B^{(1)}(n_1n_2+n_1'n_2')+B^{(2)}n_1n_2']+\cdots, \tag{39}$$

with

$$n_k=\alpha_k^*\alpha_k, \quad n_k'=\beta_k^*\beta_k, \tag{40}$$

$$E_h=2z|J|S\left\{-\frac{1}{2}SN(1-h_1'+h_2') +\sum_k[(D^2-\gamma_k^2)^{\frac{1}{2}}-D] -\frac{1}{2SN}\left[\sum_k\left(\frac{D-\gamma_k^2}{(D^2-\gamma_k^2)^{\frac{1}{2}}}-1\right)\right]^2\right\}, \tag{41}$$

$$A_k^\pm=(D^2-\gamma_k^2)^{\frac{1}{2}}\pm\tfrac{1}{2}(h_1'+h_2') -\frac{1}{SN}\frac{D-\gamma_k^2}{(D^2-\gamma_k^2)^{\frac{1}{2}}}\sum_\kappa\left(\frac{D-\gamma_k^2}{(D^2-\gamma_k^2)^{\frac{1}{2}}}-1\right), \tag{42}$$

$$B^{(1)}=-\frac{z|J|}{N}\left(\frac{D-\gamma_1^2}{(D^2-\gamma_1^2)^{\frac{1}{2}}}\frac{D-\gamma_2^2}{(D^2-\gamma_2^2)^{\frac{1}{2}}}-1\right), \tag{43}$$

$$B^{(2)}=-\frac{2z|J|}{N}\left(\frac{D-\gamma_1^2}{(D^2-\gamma_1^2)^{\frac{1}{2}}}\frac{D-\gamma_2^2}{(D^2-\gamma_2^2)^{\frac{1}{2}}}+1\right), \tag{44}$$

$$D=1-\frac{h_1'}{2}+\frac{h_2'}{2}, \tag{45}$$

where we have picked up all terms to order $(1/S^0)$. By a method similar to that of the last section, we obtain the free energy as follows:

$$F=E_h+\kappa T\sum_k \{\ln[1-\exp(-\beta A_k^-)] +\ln[1-\exp(-\beta A_k^+)]\}+\sum_{k_1,k_2} \times\left[B^{(1)}\left(\frac{1}{\exp(\beta A_1^-)-1}\frac{1}{\exp(\beta A_2^-)-1} +\frac{1}{\exp(\beta A_1^+)-1}\frac{1}{\exp(\beta A_2^+)-1}\right) +B^{(2)}\frac{1}{\exp(\beta A_1^-)-1}\frac{1}{\exp(\beta A_2^+)-1}\right]. \tag{46}$$

TABLE I. Numerical quantities for antiferromagnetic lattices.[a]

	NaCl-type	CsCl-type
c	0.097	0.073
c'	0.156	0.150
η	$1/(3^{\frac{1}{2}}2^{\frac{1}{3}})$	$\frac{1}{2}$
ξ_1	$2^{4/3}$	3
ξ_2	$39/2^{4/3}$	$87/4$

[a] The numerical values of c and c' are calculated by Anderson and by Kubo.

When the external magnetic field is zero (i.e., $h_1=h_2=0$, $D=1$) the free energy F_0 can be written,

$$F_0=E_0+2\kappa T\sum_k \ln[1-\exp(-\beta A_k^0)] -\frac{4z|J|}{N}\left(\sum_k\frac{(1-\gamma_k^2)^{\frac{1}{2}}}{\exp(\beta A_k^0)-1}\right)^2, \tag{47}$$

where A_k^0 means $A_k^-(h_1'=h_2'=0)=A_k^+(h_1'=h_2'=0)$. The zero-point energy E_0 is

$$E_0=-z|J|SN(S+c+c^2/4S), \tag{48}$$

where c is defined by

$$c=(2/N)\textstyle\sum_k[1-(1-\gamma_\kappa^2)^{\frac{1}{2}}]. \tag{49}$$

The numerical values of c are calculated by Anderson and by Kubo, and shown in Table I. Equation (48) is identical with the result of Kubo in his second approximation. As shown in Table I, c is so small that the correction term $c^2/4S$ is not important. The energy of the spin wave with wave number k is

$$A_k^0=2z|J|S(1+c/2S)(1-\gamma_k^2)^{\frac{1}{2}}. \tag{50}$$

In Eq. (50), though $c/2S$ is a correction, the property that the spin-wave energy is a linear function of k in the region of long wavelength does not change. From Eq. (47) we obtain the internal energy by the standard method:

$$U=E_0+2z|J|SN\left[\frac{3\zeta(4)}{\pi^2\eta^3}\left(1-\frac{3c}{2S}\right)\theta^4 +\frac{1}{S}\frac{63}{2}\frac{\zeta(4)^2}{\pi^4\eta^6}\theta^8+\cdots+5\xi_1\frac{\zeta(6)}{\pi^2\eta^5}\left(1-\frac{5c}{2S}\right)\theta^6 +7\xi_2\frac{\zeta(8)}{\pi^2\eta^7}\left(1-\frac{7c}{2S}\right)\theta^8+\cdots\right], \tag{51}$$

with

$$\theta=\kappa T/2z|J|S, \tag{52}$$

$$\zeta(2)=\pi^2/6, \quad \zeta(4)=\pi^4/90, \quad \zeta(6)=\pi^6/945, \quad \zeta(8)=\pi^8/9450. \tag{53}$$

In Eq. (51) the second term in the brace is the correction due to the spin-wave interactions, and the last two terms are due to corrections resulting from the deviation from linearity of the spin-wave energy spectrum. The numerical constants η, ξ_1, and ξ_2 are shown in Table I for NaCl and CsCl type crystals. The specific heat is easily obtained:

$$\frac{C_v}{N\kappa}=\frac{12\zeta(4)}{\pi^2\eta^3}\left(1-\frac{3c}{2S}\right)\theta^3+\frac{1}{S}252\frac{\zeta(4)^2}{\pi^4\eta^6}\theta^7+\cdots$$
$$+30\xi_1\frac{\zeta(6)}{\pi^2\eta^5}\left(1-\frac{5c}{2S}\right)\theta^5$$
$$+56\xi_2\frac{\zeta(8)}{\pi^2\eta^7}\left(1-\frac{7c}{2S}\right)\theta^7+\cdots. \quad (54)$$

We can understand the meaning of each term in Eq. (54) by comparing with those in Eq. (51).

In order to get the magnetization, M, of sublattice, we differentiate Eq. (46) with respect to the selector parameter h_1, using the following property:

$$\frac{\partial}{\partial h_1}\left(\frac{D-\gamma_k^2}{(D^2-\gamma_k^2)^{\frac{1}{2}}}\right)_{h_1=h_2=0}=0. \quad (55)$$

The result is

$$\frac{M}{g\mu}=\frac{N}{2}\left[S-\tfrac{1}{2}c'-\frac{\zeta(2)}{2\pi^2\eta^3}\left(1-\frac{c}{S}\right)\theta^2-\frac{1}{S}\frac{3\zeta(2)\zeta(4)}{\pi^4\eta^6}\theta^6\cdots\right.$$
$$\left.-\xi_1\frac{\zeta(4)}{4\pi^2\eta^5}\left(1-\frac{2c}{S}\right)\theta^4-\xi_2\frac{\zeta(6)}{6\pi^2\eta^7}\left(1-\frac{3c}{S}\right)\theta^6\cdots\right], \quad (56)$$

where c' is defined by

$$c'=\frac{2}{N}\sum_k\left(\frac{1}{(1-\gamma_k^2)^{\frac{1}{2}}}-1\right), \quad (57)$$

and its numerical values, as calculated by Anderson and Kubo, are shown in Table I. The magnitude of magnetization at zero temperature, $\theta=0$, does not change from Kubo's result, although Fisher[13] very recently got a different result by a variational method. The fourth term proportional to θ^6 in Eq. (56) comes from the spin-wave interactions. The last two terms are due to the energy spectrum correction.

To obtain the parallel susceptibility, $\chi_{\parallel}$, we must put $h_1=h_2=h$, and calculate the second derivative of the free energy with respect to h. The result obtained is

$$\chi_{\parallel}=\frac{N(g\mu)^2}{2z|J|S}\left[\frac{\zeta(2)}{\pi^2\eta^3}\left(1-\frac{3c}{2S}\right)\theta^2-\frac{1}{S}\frac{[\zeta(2)]^2}{\pi^4\eta^6}\theta^4\right.$$
$$+\frac{1}{S}\frac{9\zeta(2)\zeta(4)}{\pi^4\eta^6}\theta^6+\cdots+\xi_1\frac{\zeta(4)}{\pi^2\eta^5}\left(1-\frac{5c}{2S}\right)\theta^4$$
$$\left.+\xi_2\frac{\zeta(6)}{\pi^2\eta^7}\left(1-\frac{7c}{2S}\right)\theta^6-\frac{1}{S}\frac{2\xi_1}{\pi^4\eta^8}\zeta(2)\zeta(4)\theta^6\cdots\right]. \quad (58)$$

Both the second and third terms are caused by the spin-wave interactions, and are order of $1/S$.

Lastly, we will get the perpendicular susceptibility, $\chi_{\perp}$. In Eq. (31) we add the following Hamiltonian, and put h_1 and h_2 equal to zero:

$$h_x\{\sum_l S_l^x+\sum_m S_m^x\}. \quad (59)$$

This is the Hamiltonian of the external magnetic field directed along the x axis. We can use transformations similar to those in Eqs. (32) through (37). Noting that the perpendicular magnetic field shifts the origin of the operators a_0 and a_0^* by $-h_xN^{\frac{1}{2}}/4z|J|S^{\frac{1}{2}}$ while b_0 and b_0^* do not change, the Hamiltonian can be diagonalized easily within the range of the same approximation, and the perpendicular susceptibility is obtained as follows:

$$\chi_{\perp}=\frac{N(g\mu)^2}{4z|J|}\left[1-\frac{1}{2S}(c+c')-\frac{1}{S}\frac{\zeta(2)}{2\pi^2\eta^3}\left(1-\frac{c}{S}\right)\theta^2\right.$$
$$+\frac{1}{S}\frac{3\zeta(4)}{\pi^2\eta^3}\left(1-\frac{2c}{S}\right)\theta^4-\frac{1}{S^2}\frac{3\zeta(2)\zeta(4)}{\pi^4\eta^6}\theta^6\cdots$$
$$-\frac{1}{S}\xi_1\frac{\zeta(4)}{4\pi^2\eta^5}\left(1-\frac{2c}{S}\right)\theta^4+\frac{1}{S}\left(5\xi_1-\frac{\xi_2}{6\eta^2}\right)$$
$$\left.\times\frac{\zeta(6)}{\pi^2\eta^5}\left(1-\frac{3c}{S}\right)\theta^6\cdots\right]. \quad (60)$$

In this expression the third, fourth, and fifth terms represent the effect of interactions between spin waves, and the last two terms are due to the energy spectrum correction as before. Ziman[14] calculated the correction terms of the perpendicular susceptibility due to spin-wave interactions, but he did not obtain the temperature-dependent term because of his poor approximation.

All the correction terms which we obtain are quite small so that they are hardly observable in a real antiferromagnet. Nevertheless, the smallness of the effect of spin-wave interactions confirms the usefulness of the first approximation in H-P's method. In a case

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[13] J. C. Fisher, J. Phys. Chem. Solids **10**, 44 (1959). His result is $M(0)/g\mu=\frac{1}{2}NS[1-zS/(2zS-1)^2]=\frac{1}{2}NS\times0.987$ (in the case of MnF$_2$), and he compares it with the experimental result $\frac{1}{2}NS\times0.98\leqslant M(0)/g\mu\leqslant\frac{1}{2}NS\times1.03$ by Jaccarino in MnF$_2$. On the other hand Eq. (56) gives $M(0)/g\mu=\frac{1}{2}NS\times0.97$. In both theories the anisotropy energy is neglected and only the single exchange integral is considered. (If the theories are applied to MnF$_2$, this exchange integral corresponds to that between second nearest neighbors.) However we cannot neglect the effect of the anisotropy energy and the exchange integral between nearest neighbors on the magnetization in a real MnF$_2$ crystal. Thus it is difficult to say which theory is in better agreement with the experiment.

[14] J. M. Ziman, Proc. Phys. Soc. (London) **65**, 540 and 548 (1952).

in which the first approximation does not fit the experimental result, we must seek the origin in the difference between the real antiferromagnet and the simple Heisenberg model.

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Positron Annihilation in Metals

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A correlation function for a positron-electron pair within a metal is obtained by solving a Bethe-Goldstone equation. Thus one may take account of the many body effects, of screening and the exclusion principle, in computing the electron density at the position of the positron. The results indicate that one may, in this fashion, reconcile the Sommerfeld model of a metal with the experimental data on positron annihilation in metals.

I. INTRODUCTION

In recent years attempts have been made to explain the failure of the Sommerfeld model of a metal to account for the observed positron annihilation rates in metals. It was apparent that no progress could be made until some account was taken of the interactions between valence electrons and positron within the metal. We propose to present an adequate treatment of these interactions. Our work follows most closely that of Ferrell.[1] The basis of the latter's discussion was the Bohm and Pines theory[2] of collective motion in an electron gas. The introduction of the collective or plasmon modes allows one to effect a separation between the long range and short range, or screened, Coulomb interactions in the electron gas. In the present work we will make no attempt to calculate the effect on the positron annihilation rate of the plasmon part of the Hamiltonian. This will be discussed in a later paper. For now we only refer to the earlier work of Ferrell, whose estimate of the plasmon effects indicate they are probably small.

Where we differ from Ferrell is in the treatment of the screened electron-positron Coulomb force. Ferrell reasoned that this weakened interaction should admit of a perturbation treatment. Our calculations indicate this was not a justifiable assumption, at least not for most of the metals of interest. We have attempted to arrive at an electron-positron correlation function by setting down a two-body equation embodying the screened Coulomb positron-electron force and the equally important Pauli exclusion principle. This can be done by formulating the appropriate Bethe-Goldstone equation[3] for an electron-positron pair in a metal. No attempt is made here to include the proper energy-momentum relationship (dispersion law) for the electron and positron, resulting from the short range collisions experienced by these particles. It is assumed that both electron and positron propagate as free particles.

In Sec. II we set out in detail the basic wave equation for our problem. In Secs. III and IV an approximate solution of the equation is discussed and the results for electrons annihilating at zero tabulated. The calculation is extended to include electrons at the surface of the Fermi distribution in V and the resulting annihilation lifetimes computed.

II. BETHE-GOLDSTONE EQUATION FOR ELECTRON-POSITRON PAIRS

We are of course ignoring lattice effects and treating the metal as an electron gas (at zero temperature) with the neutralizing charge of the positive ions smeared throughout space. We assume that the positron on entering the metal is rapidly thermalized[4] and assign zero momentum to this particle. It is convenient to carry out our calculations entirely in momentum space, for it is then possible to state both the Pauli principle and the screening as algebraic restrictions on the intermediate relative momenta. It is easiest to set down the two-body equation we have in mind and explain the notation after.

The equation describing the interaction of a zero momentum positron with a representative electron of momentum $\mathbf{k}_e = 2\mathbf{a}$ is

$$(k^2-a^2)\phi(\mathbf{k})=\frac{4\pi}{(2\pi)^3 a_0}\int' \frac{d^3k'}{|\mathbf{k}-\mathbf{k}'|^2}\phi(\mathbf{k}'), \tag{1}$$

where $\phi(\mathbf{k})$ is the relative-momentum wave function of

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[1] R. A. Ferrell, Revs. Modern Phys. **28**, 308 (1956).

[2] D. Bohm and D. Pines, Phys. Rev. **92**, 609 (1953).

[3] H. A. Bethe and J. Goldstone, Proc. Roy. Soc. (London) **A238**, 551 (1957).

[4] G. E. Lee-Whiting, Phys. Rev. **97**, 1557 (1955).