Pseudodipolar Anisotropy in Cubic Ferromagnets at Low Temperatures*

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The first order anisotropy constant, K_1, of a cubic ferromagnet with spin 1/2 per atom is calculated as a function of temperature at low temperatures. The source of this anisotropy is taken to be the nearest neighbor pseudodipolar spin-spin interaction and the spin-wave approach of Dyson is used. It is shown that K_1 varies as the tenth power of the magnetization, itself a function of the temperature. In order to explain the experimental value of K_1 for nickel at $T=0$ the strength of the dipolar interaction must be $\sim$300 times the classical value. Previous calculations by Van Vleck, Van Peipe, and Tessman are compared with the present work on the ground state. Only the work of Van Peipe accounts properly for the exchange and is in complete agreement with the present investigation. The perturbation scheme of Van Peipe is shown to be rigorously correct, the wave function converging to an exponential form.

I. INTRODUCTION

THE free energy per unit volume of a ferromagnet contains terms which depend upon the orientation of the magnetization vector with respect to the crystalline axes. These terms are commonly distinguished by the angular dependence associated with each of them. In the case of cubic symmetry the leading term is $K_1(\alpha_1^2\alpha_2^2+\alpha_1^2\alpha_3^2+\alpha_2^2\alpha_3^2)$, where K_1 is known as the first order anisotropy constant and α_1, α_2, and α_3 are the direction cosines of the magnetization with respect to the crystalline axes. The next term is $K_2\alpha_1^2\alpha_2^2\alpha_3^2$, where K_2 is the second order anisotropy constant.

There have been several calculations of the first order anisotropy at $T=0$ of cubic ferromagnetics with spin value $\frac{1}{2}$ per atom using the short-range pseudodipolar coupling. The approach used by Van Vleck[1] is admittedly approximate in that the exchange energy is replaced by the Weiss molecular field. Tessman[2] treated the exchange by the usual spin-wave approximation and was able to show that the anisotropy is due to the fluctuations of the zero-point energy. In the explicit calculation of the anisotropy, he replaces the exchange energy by JZ, the maximum spin-wave energy, with results which agree with those of Van Vleck. It has been pointed out[3] that although the methods are different, the results are in agreement because the approximations made are equivalent to each other. The most extensive treatment of the problem was given by Van Peipe[4] who attempted a rigorous treatment of exchange. He obtained anisotropy energies somewhat larger than the others. Moreover doubt has been cast on this work[5] in

that he apparently used a perturbation expansion in powers of N, the number of atoms in the crystal, so that the convergence of the expansion is dubious.

In Sec. II we transform the Hamiltonian, including the pseudodipolar coupling, into the language of second quantization. In Sec. III the low-lying energy levels of the ferromagnet, to second order in the dipolar term, are found. These calculations are based on the spin-wave method due to Dyson.[6] The first order anisotropy constant at $T=0$, K_{10}, is immediately extracted from the ground-state energy. The anisotropic part of the free energy is calculated in Sec. IV to give K_1 as a function of the temperature. Although we do not repeat the argument here,[1,3] we point out that a dipolar interaction is not capable of giving anisotropy in a cubic crystal below the second order of perturbation. This is in contrast to the quadrupolar interaction, possible only for $S \geq 1$, which yields anisotropy in the first order perturbation. Our results agree at $T=0$ with those of Van Peipe and we point out, in Sec. III, that the other calculations differ because of their approximate treatment of the exchange. We find that K_1/K_{10} varies as the tenth power of the magnetization in the $T^{\frac{3}{2}}$ law region for all the cubic lattices.

In Sec. V we review the work of Van Peipe in atomic spin space. The treatment is extended to give an approximate ground-state wave function, appropriate for the calculation of energy to second order only. This wave function is shown to be exponential in form and the power series expansion of this wave function agrees in first order with the wave function which Van Peipe used to get the second order energy. We conclude that the Van Peipe calculation is correct since the wave function does converge. Also included in this section is a calculation of the fractional deviation of the ground-state magnetization from saturation due to the dipolar coupling for a simple cubic lattice. For this purpose the entire exponential wave function must be used and the resulting deviation is small. If the Van Peipe wave function, i.e., only the first term in the expansion is used, the result is much larger.

[1] J. H. Van Vleck, Phys. Rev. **52**, 1178 (1937).
[2] J. R. Tessman, Phys. Rev. **96**, 1192 (1954).
[3] J. Van Kranendonk and J. H. Van Vleck, Revs. Modern Phys. **30**, 1 (1958).
[4] W. F. Van Peipe, Physica **5**, 465 (1938).
[5] P. Argyres and C. Kittel, Acta Met. **1**, 241 (1953).
[6] F. J. Dyson, Phys. Rev. **102**, 1217 (1956).

The application of the results of this investigation to real ferromagnets is discussed in Sec. VI.

II. THE HAMILTONIAN

If we neglect the magnetic dipolar coupling, the spin Hamiltonian is, for $S=\frac{1}{2}$,

$$\mathcal{H}= -g\mu_B H \sum_i S_{R_i}{}^z - J \sum_{i,1} \mathbf{S}_{R_i} \cdot \mathbf{S}_{R_i+1} + 2\epsilon \sum_{i,1}(\mathbf{l}\cdot \mathbf{S}_{R_i})(\mathbf{l}\cdot \mathbf{S}_{R_i+1}). \quad (1)$$

The first term is the Zeeman energy, with the z direction being that of the applied field H. The second term is the exchange energy and the last represents the pseudodipolar coupling which gives rise to the anisotropy. The atom located at $\mathbf{R}_i$ has spin angular momentum $\hbar\mathbf{S}_{R_i}$ and the magnetic moment $g\mu_B\mathbf{S}_{R_i}$. The sums over i range over the entire crystal of N atoms. The sums on $\mathbf{l}$ range over the Z nearest neighbor vectors of the lattice, where $\mathbf{l}$ is measured in units of the nearest neighbor distance. The parameters J and ϵ measure the strengths of the exchange and pseudodipolar interactions, respectively. In particular, they have been chosen to correspond to the notation used by Van Peipe.[4] For nickel, it will be seen that $\epsilon/J\approx0.09$.

We separate the Hamiltonian into

$$\mathcal{H}=\mathcal{H}_0+\mathfrak{U}+\mathfrak{V}, \quad (2)$$

where

$$\mathcal{H}_0 = -g\mu_B H \sum_i S_{R_i}{}^z - J \sum_{i,1} \mathbf{S}_{R_i} \cdot \mathbf{S}_{R_i+1} + 2\epsilon \sum_{i,1}[(l^z)^2 S_{R_i}{}^z S_{R_i+1}{}^z + \tfrac{1}{2}(l^+l^-)S_{R_i}{}^- S_{R_i+1}{}^+],$$

$$\mathfrak{U}=2\epsilon \sum_{i,1}[(l^z l^+)S_{R_i}{}^z S_{R_i+1}{}^- + (l^z l^-)S_{R_i}{}^z S_{R_i+1}{}^+],$$

$$\mathfrak{V}=\tfrac{1}{2}\epsilon \sum_{i,1}[(l^+)^2 S_{R_i}{}^- S_{R_i+1}{}^- + (l^-)^2 S_{R_i}{}^+ S_{R_i+1}{}^+].$$

Following the usual definitions,

$$S_{R_i}{}^{\pm}=S_{R_i}{}^x \pm i S_{R_i}{}^y \quad \text{and} \quad l^{\pm}=l^x \pm il^y. \quad (3)$$

The dc part of the dipolar interaction has been included in $\mathcal{H}_0$; that part which gives rise to the first order anisotropy at $T=0$ is $\mathfrak{V}$, and $\mathfrak{U}$ contributes to K_1 in the excited states only. At $T=0$, however, the $\mathfrak{U}$ term must be included in a calculation of K_2, the second order anisotropy constant.

We introduce the spin-wave operators,

$$\mathbf{S}_k=N^{-\frac{1}{2}}\sum_i \mathbf{S}_{R_i}e^{-i\mathbf{k}\cdot\mathbf{R}_i}. \quad (4)$$

They obey the commutation rules

$$[S_k{}^z,S_{k'}{}^{\pm}]=\pm N^{-\frac{1}{2}}S_{k+k'}{}^{\pm} \quad \text{and}$$
$$[S_k{}^+,S_{k'}{}^-]=2N^{-\frac{1}{2}}S_{k+k'}{}^z. \quad (5)$$

In terms of these operators

$$\mathcal{H}_0 = -g\mu_B H N^{\frac{1}{2}}S_0{}^z - J \sum_{k,1} \exp(-i\mathbf{k}\cdot\mathbf{l}) \times (S_k{}^z S_{-k}{}^z + S_k{}^- S_{-k}{}^+) + 2\epsilon \sum_{k,1} \exp(-i\mathbf{k}\cdot\mathbf{l}) \times [(l^z)^2 S_k{}^z S_{-k}{}^z + \tfrac{1}{2}(l^+l^-)S_k{}^- S_{-k}{}^+], \quad (6a)$$

$$\mathfrak{U}=2\epsilon \sum_{k,1} \exp(-i\mathbf{k}\cdot\mathbf{l}) \times [(l^z l^+)S_k{}^z S_{-k}{}^- + (l^z l^-)S_k{}^z S_{-k}{}^+], \quad (6b)$$

$$\mathfrak{V}=\tfrac{1}{2}\epsilon \sum_{k,1} \exp(-i\mathbf{k}\cdot\mathbf{l}) \times [(l^+)^2 S_k{}^- S_{-k}{}^- + (l^-)^2 S_k{}^+ S_{-k}{}^+]. \quad (6c)$$

A spin-wave state in which $n(=\sum_k n_k)$ spin waves are excited is denoted by

$$|n\rangle=\prod_k(n_k!)^{-\frac{1}{2}}(S_k{}^-)^{n_k}|0\rangle, \quad (7)$$

where $|0\rangle$ represents the ground state of $\mathcal{H}_0$, the state with $n=0$. Except for terms of order n/N, these states are normalized and orthogonal.

An approximate Hamiltonian will be written in terms of the ordinary creation and destruction operators, $A_k{}^*$ and A_k, which operate on spin waves. To construct this Hamiltonian we let $\mathcal{H}$ operate on the wave function (7). The operators $S_k{}^z$ and $S_k{}^+$ are commuted through the $S_k{}^-$ operators in $|n\rangle$ until they operate on $|0\rangle$. Then use is made of $S_k{}^+|0\rangle=0$ and $S_k{}^z|0\rangle=SN^{\frac{1}{2}}|0\rangle\delta_{k,0}$. At this stage, $S_k{}^-$ is replaced by $A_k{}^*$. The original Hamiltonian is replaced by one expressed in terms of $A_k{}^*$ and A_k which, when operating on (7) with $A_k{}^*$ substituted for $S_k{}^-$, yields the same linear combination of spin-wave functions. This procedure has already been carried out by Dyson[6] for the Zeeman and exchange terms. As an example we treat $\mathfrak{U}$ here. By application of the commutation rules,

$$S_k{}^z S_{-k}{}^- S_{k_1}{}^- S_{k_2}{}^- \cdots S_{k_n}{}^-|0\rangle$$
$$= S_{-k}{}^- S_{k_1}{}^- S_{k_2}{}^- \cdots S_{k_n}{}^- S_k{}^z|0\rangle$$
$$\quad - N^{-\frac{1}{2}}\{S_0{}^- S_{k_1}{}^- S_{k_2}{}^- \cdots S_{k_n}{}^-$$
$$\quad + S_{-k}{}^- S_{k_1+k}S_{k_2}{}^- \cdots S_{k_n}{}^- + \cdots$$
$$\quad\quad\quad + S_{-k}{}^- S_{k_1}{}^- S_{k_2}{}^- \cdots S_{k_n+k}{}^-\}|0\rangle.$$

The term containing $S_k{}^z$ makes no contribution to $\mathfrak{U}|n\rangle$ since only the $\mathbf{k}=0$ part survives, and $\sum_1(l^z l^+)=0$ for cubic symmetry. The next term also vanishes since we may remove as a factor $\sum_k \exp(i\mathbf{k}\cdot\mathbf{l})=0$. The remaining terms have the following interpretation; one of the spin waves, say $\mathbf{k}_i$, is destroyed and two others, $-\mathbf{k}$ and $\mathbf{k}_i+\mathbf{k}$ are created in its place. The $S_k{}^-$ are clearly equivalent to creation operators, so that the $S^z S^-$ portion of $\mathfrak{U}$ may be replaced by

$$-\epsilon N^{-\frac{1}{2}} \sum_{k,k'} (u_k+u_{k'})A_k{}^* A_{k'}{}^* A_{k+k'},$$

where

$$u_k=\sum_1(l^z l^+) \exp(i\mathbf{k}\cdot\mathbf{l}).$$

By following a similar procedure, the Hermitian adjoint is obtained for the $S^z S^+$ portion.

The Hamiltonian in terms of the creation and destruction operators is,

$$\mathcal{H}=\mathcal{H}^0+\mathcal{H}'+\mathfrak{U}+\mathfrak{V}, \quad (8)$$

where

$$\mathcal{H}^0=C+\sum_k n_k\epsilon_k,$$

$$\mathfrak{K}' = -\tfrac{1}{2}N^{-1}J \sum_{\mathbf{k}} \sum_{\mathbf{k}',\mathbf{k}''} \Gamma_{\mathbf{k}'\mathbf{k}''}{}^{\mathbf{k}} A_{\mathbf{k}'+\mathbf{k}}{}^{*} A_{\mathbf{k}''-\mathbf{k}}{}^{*} A_{\mathbf{k}'} A_{\mathbf{k}''},$$

$$\mathfrak{U} = -\epsilon N^{-\frac{1}{2}} \sum_{\mathbf{k},\mathbf{k}'} \big[(u_{\mathbf{k}}{}^{*}+u_{\mathbf{k}'}{}^{*}) A_{\mathbf{k}+\mathbf{k}'}{}^{*} A_{\mathbf{k}} A_{\mathbf{k}'}$$
$$+ (u_{\mathbf{k}}+u_{\mathbf{k}'}) A_{\mathbf{k}}{}^{*} A_{\mathbf{k}'}{}^{*} A_{\mathbf{k}+\mathbf{k}'} \big],$$

$$\mathfrak{V} = \tfrac{1}{2}\epsilon \sum_{\mathbf{k}} (v_{\mathbf{k}} A_{\mathbf{k}}{}^{*} A_{-\mathbf{k}}{}^{*} + v_{\mathbf{k}}{}^{*} A_{\mathbf{k}} A_{-\mathbf{k}})$$
$$- (\epsilon/N) \sum_{\mathbf{k},\mathbf{k}',\mathbf{k}''} v_{\mathbf{k}}{}^{*} A_{\mathbf{k}+\mathbf{k}'+\mathbf{k}''}{}^{*} A_{\mathbf{k}} A_{\mathbf{k}'} A_{\mathbf{k}''}.$$

Here C is a constant dependent upon the lattice and given by

$$C = -\tfrac{1}{2}g\mu_B N H - \tfrac{1}{4}NJZ + \tfrac{1}{2}N\epsilon \sum_1 (l^z)^2. \quad (9)$$

The energy of each spin wave is, to first order,

$$\epsilon_{\mathbf{k}} = g\mu_B H + J \sum_1 (1-\cos\mathbf{k}\cdot\mathbf{l})$$
$$- \epsilon \sum_1 [2(l^z)^2 - (l^+l^-)\cos\mathbf{k}\cdot\mathbf{l}]. \quad (10)$$

The portion $\mathfrak{K}'$, like $\mathfrak{K}^0$, is derived from $\mathfrak{K}_0$. It gives rise to binary collisions between spin waves with

$$\Gamma_{\mathbf{k}_1\mathbf{k}_2}{}^{\mathbf{k}} = \sum_1 \exp(i\mathbf{k}\cdot\mathbf{l})[1-\exp(i\mathbf{k}_1\cdot\mathbf{l})]$$
$$\times [1-\exp(-i\mathbf{k}_2\cdot\mathbf{l})]. \quad (11)$$

We have also

$$v_{\mathbf{k}} = \sum_1 (l^+)^2 \exp(i\mathbf{k}\cdot\mathbf{l}).$$

There are terms of higher order in $1/N$ belonging to $\mathfrak{U}$ and $\mathfrak{V}$ which are not shown. These give rise to contributions to the energy which, for the low-lying states, are negligible.[7] The specific advantage of the procedure outlined above is in the interaction terms $\mathfrak{K}'$ and $\mathfrak{V}$, which are different from the corresponding terms in the usual spin-wave theory.[8] As derived here these terms are the complete effective spin-wave interaction which is contained in Eqs. (6a) and (6c).

It may be observed that $\mathfrak{K}^0 + \mathfrak{V}$ (except for the term proportional to ϵN^{-1}) is the Hamiltonian derived by Holstein and Primakoff[8] with the long-range magnetic dipole forces replaced by the pseudodipolar coupling. This Hamiltonian was diagonalized by them by means of a canonical transformation. We have obtained their result for the ground-state energy by regarding $\mathfrak{V}$ as a perturbation on $\mathfrak{K}^0$ and summing the infinite series as was done by Brueckner and Sawada[9] in connection with the problem of liquid helium. The series corresponds to the binomial expansion of the square root in the Holstein and Primakoff result. The second order term in $\mathfrak{V}$ is just the one used by Tessman[2] to obtain the first order anisotropy of the ground state. This procedure of evaluating and summing the infinite series is not included here.[10]

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[7] The last term in $\mathfrak{V}$ was originally included in this class by us. Neglect of this term led to an eighth power law and to an instability of the spin-wave spectrum. The importance of this term was pointed out by F. Keffer (private communication).

[8] T. Holstein and H. Primakoff, Phys. Rev. **58**, 1098 (1940).

[9] K. A. Bruckner and K. Sawada, Phys. Rev. **106**, 1117 (1957).

[10] S. H. Charap, thesis, Rutgers University, 1958 (unpublished).

III. EIGENVALUES OF THE ENERGY

If $\mathfrak{K}'+\mathfrak{U}+\mathfrak{V}$ is treated as a perturbation on $\mathfrak{K}^0$, there are contributions to the energy from $\mathfrak{K}'$ alone. For the low-lying states these are negligible. The smallness of this dynamical interaction is one of the major results of Dyson's[6] work. In fact all contributions to the energy in the perturbation series in which matrix elements of $\mathfrak{K}'$ appear connecting the unperturbed state to an intermediate state are negligible (of order n/N) compared to the terms which we shall keep. This is due to the suppression of the sums over wave vectors imposed in these cases. On the other hand, those contributions to the energy in which matrix elements which connect only intermediate states via $\mathfrak{K}'$ appear are significant. If $\mathfrak{K}'$ were the sole perturbation then, it would contribute a negligible amount to the energy. But this term has a significant influence on contributions made by $\mathfrak{U}$ and $\mathfrak{V}$. For this reason we calculate contributions to second powers in $\mathfrak{U}$ and $\mathfrak{V}$, but to all powers $\mathfrak{K}'$! Consequently the energy to second order in the pseudodipolar interaction, $\mathfrak{U}+\mathfrak{V}$, is an infinite series, each succeeding term of which involves one more collision of a pair of spin waves in the intermediate states by way of exchange ($\mathfrak{K}'$) than the previous one. The entire series may be summed. With one unimportant exception pointed out below Eq. (15), each succeeding term is just the previous one multiplied by a small numerical factor ν_j, dependent upon the type of lattice involved. This is just the geometric series. In the case of the exceptional term, the sum of the series is the derivative of the geometric series with respect to ν_j.

There are contributions to the energy of higher order in the dipolar interaction. Even if there are such terms having the angular dependence associated with K_1 these are smaller than those which are calculated here by factors of $(\epsilon/J)^2$ or smaller, and may be neglected.

The second order energy of the unperturbed state $|n\rangle$ is

$$E_2{}^n = \langle n | \mathfrak{V}a^{-1}\mathfrak{V} | n \rangle + \langle n | \mathfrak{U}a^{-1}\mathfrak{U} | n \rangle, \quad (12)$$

and the third order energy is

$$E_3{}^n = \langle n | \mathfrak{V}a^{-1}\mathfrak{K}'a^{-1}\mathfrak{V} | n \rangle + \langle n | \mathfrak{U}a^{-1}\mathfrak{K}'a^{-1}\mathfrak{U} | n \rangle. \quad (13)$$

In these expressions,[11] $a = E_0{}^n - H^0$, where

$$(E_0{}^n - \mathfrak{K}^0) | n \rangle = 0.$$

A straightforward calculation yields

$$E_2{}^n = -\frac{\epsilon^2}{4} \sum_{\mathbf{k}} \frac{|v_{\mathbf{k}}|^2}{\epsilon_{\mathbf{k}}} \left(1 + 2n_{\mathbf{k}} - \frac{4}{N} \sum_{\mathbf{k}'} n_{\mathbf{k}'} \right)$$
$$- 2\frac{\epsilon^2}{N} \sum_{\mathbf{k},\mathbf{k}'} \frac{|u_{\mathbf{k}}+u_{\mathbf{k}'}|^2 n_{\mathbf{k}+\mathbf{k}'}}{\epsilon_{\mathbf{k}}+\epsilon_{\mathbf{k}'}-\epsilon_{\mathbf{k}+\mathbf{k}'}}. \quad (14)$$

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[11] This notation for perturbation theory is standard in the theory of scattering. See, for example, J. Goldstone, Proc. Roy. Soc. (London) **A239**, 267 (1957), or K. A. Brueckner, Phys. Rev. **100**, 36 (1955), Sec. III.

In these terms products of the n_k have been neglected. It is now evident why only $\mathcal{V}$ contributes to the ground-state anisotropy; the last term, arising from the $\mathcal{U}$ portion, vanishes when no spin waves have been excited. Now $E_2{}^0$ is the term evaluated by Tessman.[2] In doing so he set $\epsilon_k = JZ$. This term, as well as all of Eq. (14), is evaluated in Appendix A without approximating the exchange energy and the values of K_{10} calculated in this way for the three cubic lattices are shown in Table I. The third order energy is more involved. In Appendix B it is shown that, if only the long wavelengths are excited,

$$E_3{}^n = -\nu_j\left\{ \frac{\epsilon^2}{4}\sum_k \frac{|v_k|^2}{\epsilon_k}\left(1+4n_k-\frac{4}{N}\sum_{k'}n_{k'}\right)\right.$$
$$\left.+\frac{2\epsilon^2}{N}\sum_{k,k'}\frac{|u_k+u_{k'}|^2 n_{k+k'}}{\epsilon_k+\epsilon_{k'}-\epsilon_{k+k'}}\right\}, \quad (15)$$

where $\nu_j = 0.211$, 0.1339, 0.08161, for the simple, body-centered and face-centered cubics, respectively. Notice that there is a term in which each spin wave is counted twice as compared to the corresponding term in E_2. In the next order each spin wave is counted three times in this term, and so on. If we sum the entire series, the energy, to second order in the pseudodipolar coupling and to all orders in $\mathcal{3C}'$ is

$$E^n = C + \sum_k n_k \epsilon_k$$
$$-(1-\nu_j)^{-1}\left\{\frac{\epsilon^2}{4}\sum_k \frac{|v_k|^2}{\epsilon_k}\left[1+2(1-\nu_j)^{-1}n_k-\frac{4}{N}\sum_{k'}n_{k'}\right]\right.$$
$$\left.+\frac{2\epsilon^2}{N}\sum_{k,k'}\frac{|u_k+u_{k'}|^2 n_{k+k'}}{\epsilon_k+\epsilon_{k'}-\epsilon_{k+k'}}\right\}. \quad (16)$$

At low temperatures only the long-wavelength spin waves are excited. In that case one finds, for Eq. (16)

$$E^n = E_g + \sum_k n_k e_k. \quad (17)$$

The ground-state energy to this order is

$$E_g(\text{sc}) = C + 0.268N(\epsilon^2/J)(\Gamma-1),$$
$$E_g(\text{bcc}) = C - 0.1374N(\epsilon^2/J)(\Gamma+1), \quad (18)$$
$$E_g(\text{fcc}) = C - 0.05430N(\epsilon^2/J)(\Gamma+2.39).$$

TABLE I. The first order anisotropy constant at $T=0$, K_{10}, in units of $\epsilon^2 N/JV$, for $S=\frac{1}{2}$, for the cubic lattices. Here $E_2{}^0$ is a refinement of Tessman's result, and $(1-\nu_j)^{-1}E_2{}^0$ takes into account the interaction between spin waves due to exchange.

	Simple cubic (sc)	Body centered cubic (bcc)	Face centered cubic (fcc)
Tessman	0.167	−0.111	−0.0419
Van Peipe	0.26(4)[a]	−0.14	−0.05±0.03
from $E_2{}^0$	0.211	−0.1190	−0.04987
from $(1-\nu_j)^{-1}E_2{}^0$	0.268	−0.1374	−0.05430

[a] Third figure calculated by authors.

The spin-wave energy may be written, (see the Appendixes),

$$e_k(\text{sc}) = \epsilon_k - 5.35(\epsilon^2/J)(\Gamma-\tfrac{1}{5}),$$
$$e_k(\text{bcc}) = \epsilon_k + 2.748(\epsilon^2/J)(\Gamma-\tfrac{1}{5}), \quad (19)$$
$$e_k(\text{fcc}) = \epsilon_k + 1.0863(\epsilon^2/J)(\Gamma-1/4.45).$$

In these expressions terms proportional to $(\epsilon^2/J)k^2$ have been neglected in comparison with the exchange energy contained in ϵ_k which is proportional to Jk^2. The angular dependence is given by the function $\Gamma = \alpha_1{}^2\alpha_2{}^2+\alpha_1{}^2\alpha_3{}^2+\alpha_2{}^2\alpha_3{}^2$ where α_1, α_2, α_3 are the direction cosines of the field relative to the crystal axes.

The first order anisotropy energy is given by $K_1 V\Gamma$, where V is the volume of the sample. The values of K_1 for the ground state, K_{10}, are given in Table I. The magnitude of the pseudodipolar coupling constant, ϵ, may be estimated by comparison with experiment. For nickel, a face-centered cubic, the experimental value of K_{10} is $\sim750\,000$ ergs/cc.[12] We will use $J\sim230K_B$ and $N/V\sim5\times10^{22}$. Here K_B is Boltzmann's constant. The numerical value of ϵ/J then turns out to be ~0.09. The strength of the classical dipolar interaction between nearest neighbors is about 300 times smaller.

The energy of a spin wave is, by Eq. (19), essentially positive. The function Γ ranges from 0 to $\frac{1}{3}$ in a way that the coefficient of ϵ^2/J is positive if the magnetization lies along an easy direction ([100] for the simple cubic, [111] for the others). If the magnetization is to be in some other direction it must be the result of the application of a magnetic field. The Zeeman energy required to excite a spin wave in such a field will always be sufficient to keep the excitation energy of a spin wave positive.

IV. TEMPERATURE DEPENDENCE OF K_1

The spin waves may, at low temperatures, be regarded as a system of noninteracting Bose particles of energy e_k. The free energy of the system is

$$F(T) = E_g + K_BT\sum_k \ln[1-\exp(-e_k/K_BT)]. \quad (21)$$

We separate the spin-wave energy into the two parts

$$e_k = e_k{}^0 + \Delta e_k, \quad (22)$$

where Δe_k contains the dependence upon the direction of the field with respect to the lattice and is small compared to $e_k{}^0$. The anisotropic part of the free energy is easily shown to be

$$(1-\nu_j)^{-1}E_2{}^0 + \sum_k\langle n_k\rangle\Delta e_k. \quad (23)$$

Here $\langle n_k\rangle$ is given by the Bose-Einstein distribution. The second term of (23) contains the temperature dependence in a series of powers of K_BT/J, the lowest

[12] R. M. Bozorth, *Ferromagnetism* (D. Van Nostrand Company, Inc., Princeton, 1951), p. 569.

power being $\frac{3}{2}$. The leading term is of the form $\alpha\Gamma\sum_k\langle n_k\rangle$. The other terms are negligible in the temperature region under consideration. The anisotropy to to first order is given by

$$K_1=K_{10}+(\alpha/V)\sum_k\langle n_k\rangle. \qquad (24)$$

In terms of the magnetization, $M(T)$, it may be shown that, in the $T^{\frac{3}{2}}$ law region for $M(T)$,

$$\frac{K_1}{K_{10}}=\left[\frac{M(T)}{M_0}\right]^{-\alpha NS/K_{10}V}, \qquad (25)$$

where M_0 is the saturation value of the magnetization. The value of the exponent $-(\alpha NS/K_{10}V)$ is 10 for all cubic lattices, independent of the evaluation of the integrals for α and K_{10}, themselves!

V. THE GROUND STATE

In this section we treat the ferromagnetic ground state in order to answer the question raised by Argyres and Kittel[13] concerning the convergence of the perturbation series used by Van Peipe,[4] in calculating K_{10}. It is asserted that, because the expansion is apparently in powers of N, the results are not rigorous. We show that, although the first order wave function is a poor approximation to the ground state, it is part of a convergent series. The use of this wave function to evaluate the second order energy is thereby justified.

As is pointed out in Appendix A, in obtaining Eq. (A2), the anisotropy of the energy of the state of complete alignment of the spins, the ground state, is insensitive to the strength of the applied field. For the purposes of this section then, a limited Hamiltonian may be used, consisting of the exchange energy plus the $\mathcal{U}$ portion of the pseudodipolar coupling. The dc part of the dipolar term is neglected along with the Zeeman energy. Since we are mainly concerned with convergence, we consider in each order in N only the terms to lowest power in ϵ/J. Thus we drop the $\mathcal{U}$ portion of the dipolar term and calculate the wave function using only the first term of (6c) as the perturbation.

We recall that in the spin-wave treatment, except for the collision part of the exchange, the corrections to the wave function due to $\mathcal{U}$ correspond to the excitation of independent pairs of spin waves of wave vectors $\mathbf{k}$ and $-\mathbf{k}$. This leads us to try, as an approximate wave function

$$\Psi=e^g\psi_0, \qquad (26)$$

where ψ_0 represents the unperturbed wave function of the ground state and g is an operator bilinear in $S_{R_i}^-$. The expansion in powers of g is assumed to correspond

to the ordinary perturbation series. Thus

$$\Psi=\psi_0+\psi_1+\psi_2+\cdots,$$

with $\psi_1=g\psi_0$, $\psi_2=(2!)^{-1}g^2\psi_0$, and so on. The operator g will be determined so as to make ψ_1 the correct first order wave function. In the higher orders if ψ_n' is the proper nth order wave function, we write $\psi_n'=\psi_n+\varphi_n$, where φ_n is a correction which must be small if the exponential form is to be a good approximation to the ground-state wave function. In the limit $N\to\infty$ we show this to be true, for φ_n is found to be of order n/N compared to ψ_n, and can therefore be neglected. However, if N is finite, when n is comparable to N, φ_n may not be neglected in comparison with ψ_n. The exponential wave function is nevertheless a good approximation to the ground state in this case also, for the following reason; in the case of spin $\frac{1}{2}$ per atom, there cannot be more than one spin reversed on any one atomic site, and the series does not increase in powers of N throughout. At first succeeding terms are N times as large as their predecessors, but when the number of terms becomes comparable to N, succeeding terms become smaller by factors of ϵ/J. Thus, for those values of n ($n\sim N$) for which $\varphi_n\sim\psi_n$, one may neglect ψ_n itself in comparison with the preceding terms in the wave function, because this term is ϵ/J smaller than the previous one, not $N(\epsilon/J)$!

By standard perturbation theory, the corrections to the energy are given by

$$E_n=(\psi_0,\mathcal{U}\psi_{n-1}'), \qquad (27)$$

where

$$(E_0-\mathcal{H}_0)\psi_0=0, \qquad (28a)$$

$$(E_0-\mathcal{H}_0)\psi_1'=\mathcal{U}\psi_0, \qquad (28b)$$

$$(E_0-\mathcal{H}_0)\psi_2'=\mathcal{U}\psi_1'-E_2\psi_0, \quad\text{and so on.} \quad (28c)$$

The unperturbed Hamiltonian, $\mathcal{H}_0$, is simply the exchange operator in the present approximation. Setting $\psi_1'=\psi_1=g\psi_0(\varphi_1=0)$, and

$$g=\frac{1}{2}\sum_{i,r}B_r S_{R_i}^- S_{R_i+r}^-, \qquad (29)$$

where $\mathbf{r}$ is summed over all the vectors of the lattice, we find by manipulation of Eq. (28b),

$$\sum_1\{J[(B_r-B_{r+1})-(B_r-B_0)\delta_{1,r}]+\tfrac{1}{2}\epsilon(l^+)^2\delta_{1,r}\}=0. \quad (30)$$

This equation corresponds exactly to Eqs. (19) of Van Peipe's[4] paper. Note that Eq. (28b) may be written $[g,\mathcal{H}_0]\psi_0=\mathcal{U}^-\psi_0$, where $\mathcal{U}$ is that part of $\mathcal{U}$ proportional to $(l^+)^2$. The B_r which solve Eq. (30) yield

$$[g,\mathcal{H}_0]=\mathcal{U}^-+\chi, \qquad (31)$$

where $\chi\psi_0=0$. However $\chi\psi_n$ is of order n compared to ψ_n. Equation (28c) may be written

$$\tfrac{1}{2}[g^2,\mathcal{H}_0]\psi_0+(E_0-\mathcal{H}_0)\varphi_2=\mathcal{U}g\psi_0-E_2\psi_0=\mathcal{U}^-g\psi_0.$$

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[13] See reference 5. The authors are grateful to Professor Elihu Abrahams, who brought this letter to their attention.

Making use of Eq. (31) and the fact that $[g,\mathcal{V}^-]=0$, we find

$$(E_0-\mathcal{3C}_0)\varphi_2=-\tfrac{1}{2}(g\chi+\chi g)\psi_0.$$

Therefore φ_2 is of order N smaller than ψ_2. Compared to ψ_1, φ_2 is of order ϵ/J smaller. The same procedure may be carried out for the higher order terms with the result that $\varphi_n \sim (n/N)\psi_n$. Thus, as discussed above, the exponential wave function is a good approximation to the ground state.

The second order energy is just

$$E_2=(\psi_0,\mathcal{V}g\psi_0)=\tfrac{1}{2}\epsilon \sum_{i,1}(l^-)^2 B_1. \tag{32}$$

This is just the quantity evaluated by Van Peipe,[4] who made use of the fact that the first order anisotropy is described by the second order energy and therefore calculated the wave function to first order only in the dipolar interaction. It has been shown here that the series does indeed converge so that Eq. (32) does yield the rigorous result.

It is interesting to calculate the fractional deviation of the ground-state magnetization from saturation. This is

$$\frac{\Delta M}{M_0}=\frac{1}{NS}\frac{(\Psi,[\sum_i S_{R_i}{}^z,e^g]\psi_0)}{(\Psi,\Psi)}=-\frac{2}{NS}\frac{(\Psi,g\Psi)}{(\Psi,\Psi)}, \tag{33}$$

since the commutator $[\sum_i S_{R_i}{}^z,g]=-2g$. If we define $\Psi(\lambda)=e^{\lambda g}\psi_0$, and note that $\partial\Psi(\lambda)/\partial\lambda=g\Psi(\lambda)$, then it may be shown that

$$\frac{\Delta M}{M_0}=-\frac{1}{NS}\frac{\partial}{\partial\lambda}\ln(\Psi(\lambda),\Psi(\lambda))|_{\lambda=1}.$$

We expand $\Psi(\lambda)$ in a power series and find on collecting terms that

$$(\Psi(\lambda),\Psi(\lambda))=\exp[\lambda^2(\psi_0,|g|^2\psi_0)].$$

Then, finally, making use of Eq. (29)

$$\Delta M/M_0=-(1/S)\sum_r|B_r|^2. \tag{34}$$

In evaluating the sum in this last equation one must be sure to include the entire lattice. If the magnetization is calculated by use of the Van Peipe wave function alone, the sum is over nearest neighbor vectors only and the result is much larger [see Eq. (36)].

In order to evaluate the B_r we make the Fourier transformation

$$B_r=\sum_k B_k e^{ik\cdot r}. \tag{35}$$

With $\delta_{1,r}=N^{-1}\sum_k \exp[ik\cdot(1-r)]$, Eq. (30) becomes

$$\sum_k\{2\epsilon_k B_k+N^{-1}\epsilon v_k-N^{-1}\sum_{k'} B_{k'}\Gamma_{k'-k'}{}^{(k-k')}\}e^{ik\cdot r}=0.$$

The last term in the braces evidently represents the

collision term denoted by $\mathcal{3C}'$ in Sec. II. Neglecting this we have

$$B_r=-\frac{\epsilon}{2N}\sum_k \frac{v_k}{\epsilon_k}e^{ik\cdot r}. \tag{36}$$

The same result may be obtained by consideration of the ground-state wave function in spin-wave language in the presence of the perturbation $\mathcal{V}$ only, and neglecting $\mathcal{3C}'$. That part containing $v_k{}^*$ may be neglected since, for any number of reversed spins its contribution is of higher order in ϵ. Compared with terms to the same order in ϵ it is of order N^{-1} or smaller. Then by ordinary perturbation methods, the ground state wave function is the following product;

$$\Psi=\prod_k\left[1-\frac{\epsilon}{2}\frac{v_k}{2\epsilon_k}A_k{}^*A_{-k}{}^*\right.$$
$$\left.+\left(\frac{\epsilon}{2}\right)^2\left(\frac{v_k}{2\epsilon_k}\right)^2(A_k{}^*A_{-k}{}^*)^2+\cdots\right]\psi_0.$$

If the product is expanded, one finds

$$\Psi=\left[1-\frac{\epsilon}{2}\sum_k \frac{v_k}{2\epsilon_k}A_k{}^*A_{-k}{}^*\right.$$
$$\left.+\frac{1}{2}\left(\frac{\epsilon}{2}\right)^2\sum_{k,k'}\frac{v_k v_{k'}}{2\epsilon_k 2\epsilon_{k'}}A_k{}^*A_{-k}{}^*A_{k'}{}^*A_{-k'}{}^*+\cdots\right]\psi_0.$$

This is just the exponential $\Psi=e^g\psi_0$, with

$$g=-\frac{\epsilon}{2}\sum_k \frac{v_k}{2\epsilon_k}A_k{}^*A_{-k}{}^*$$

$$=-\frac{\epsilon}{2N}\sum_{i,j}S_{R_i}{}^-S_{R_j}{}^-\sum_k e^{-ik\cdot(R_i-R_j)}.$$

This corresponds exactly to g as given by Eq. (29) with B_r given by Eq. (36).

If we use this value of B_r in E_2 as given by Eq. (32), the energy $E_2{}^0$ of Sec. II results. In order to calculate the exact solution of Eq. (30) we follow the method used by Van Peipe.[4]

Set

$$B_r=\sum_1 \rho_1 b_{r+1}, \tag{37}$$

where the b_r satisfy

$$\sum_1(b_r-b_{r+1})=4\pi\delta_{r,0}. \tag{38}$$

Again, the sums on 1 range over the Z nearest neighbor vectors of the lattice. Now make the Fourier transformation

$$b_r=\sum_k b_k e^{ik\cdot r}.$$

Equation (38) may be solved for the b_k and one has

$$b_k=4\pi/[N\sum_1(1-\cos k\cdot 1)],$$

and

$$b_r = \frac{4\pi}{N}\sum_k \frac{e^{ik\cdot r}}{\sum_l(1-\cos k\cdot l)}. \tag{39}$$

We substitute the expression (37) into Eq. (30), and, making use of (38) we get

$$4\pi\rho_{-1} - \sum_{l'}\rho_{l'}b_{l+l'} = -(\epsilon/2J)(l^+)^2. \tag{40}$$

By symmetry $\rho_{-1}=\rho_1$. The b_r are given for all cases of interest in Appendix A. If Eq. (40) is summed over l we find, since $\sum_l b_{l+l'}$ is independent of l' that $\sum_l\rho_l=0$. Using this fact and the technique described in Appendix A just below the listing of the b_{fgh}, we manipulate Eq. (40) to give

$$\rho_1 = -\frac{\epsilon}{2J}(l^+)^2\frac{1}{4\pi(1-\nu_j)}, \tag{41}$$

where the ν_j are defined in Appendix A. We substitute (39) and (41) in (37) to get the result

$$B_r = -(1-\nu_j)^{-1}\frac{\epsilon}{2N}\sum_k\frac{\nu_k}{\epsilon_k}e^{ik\cdot r}. \tag{42}$$

Comparison with the results of Sec. III shows that the B_r do contain the exchange collision interaction.

Now Eq. (34) becomes

$$\frac{\Delta M}{M_0} = -\frac{\epsilon^2}{2N}(1-\nu_j)^{-2}\sum_k\frac{|\nu_k|^2}{\epsilon_k^2}. \tag{43}$$

For the simple cubic lattice, with the magnetization along the [001] direction, and in the quadratic approximation for ϵ_k, it turns out that

$$\frac{\Delta M}{M_0} = -(0.79)^{-2}\frac{2}{15}\left(\frac{\epsilon}{J}\right)^2 = -0.21\left(\frac{\epsilon}{J}\right)^2. \tag{44}$$

In comparing with the result given by Argyres and Kittel[5] note that our exchange integral is theirs and that their dipolar coupling strength, C, is 4ϵ. Furthermore, in their semiclassical calculation they do not take the exchange collision term, $\mathcal{K}'$, into account. In Eq. (44) this term gives rise to the factor $(0.79)^{-2}$. For a spin of $\frac{1}{2}$, in the same circumstances, they have $-1/60(C/J)^2=-4/15(\epsilon/J)^2$, which is too large by a factor of 2.

The result of this section is simply this; the Van Peipe calculation has been shown to be a rigorously correct one on the model used. Calculation of K_1 may be made with the wave function known to first order only. A knowledge of the entire wave function is necessary in order to calculate the magnetization and other properties.

VI. CONCLUSION

The pseudodipolar interaction has been separated into a number of parts expressed in terms of Dyson's[6] spin-

wave operators. One of these is diagonal in the spin-wave states and is therefore isotropic. Another, $\mathcal{V}$, which connects states differing in spin-wave occupation numbers by two, gives rise to the first order anisotropy of the zero point energy and also contributes one-fifth of the temperature dependence of K_1 at low temperatures. The remainder of the temperature dependence is produced by the last of these parts, $\mathcal{U}$, which connects states differing in spin-wave occupation numbers by one. This portion does not, however, contribute to K_1 at $T=0$ because the cubic symmetry causes the matrix elements connecting the ground state to states having just one spin wave excited to vanish.

Keffer has argued[14] that the pseudodipolar interaction gives rise to a negligible temperature dependence of K_1. This error was due to neglect of the terms in the Holstein-Primakoff[8] Hamiltonian of order $\epsilon N^{-\frac{1}{2}}$, which do contribute significantly to the spin-wave spectrum. We, on the other hand, are indebted to Professor Keffer for pointing out the importance of the term proportional to ϵN^{-1} in our Hamiltonian, which is essential to the previously mentioned $\frac{1}{5}$ of the temperature dependence.

A calculation based on the Holstein-Primakoff formalism of the temperature dependence of K_1 due to this interaction was made by Kasuya.[15] In this calculation it was not possible to take into account the exchange interaction between spin waves, nor was it possible to get that part of the pseudodipolar interaction proportional to ϵN^{-1}. In the present work the exchange interaction is shown to give rise to corrections of as much as 25% in the energy. But the temperature dependence, expressed in terms of the magnetization, involves a ratio of energies and therefore this correction is unimportant for this purpose. However, the addition of this pseudodipolar term does affect the dependence of K_1 on the magnetization, increasing the power law from the eighth to the tenth power.

There has been no attempt made to determine the temperature at which the analysis of this paper fails because of the approximations which have been made, nor has any attempt been made to determine the temperature dependence of the pseudodipolar coupling parameter, which has here been implicitly assumed constant. Carr[16] has proposed the following empirical expression for the temperature dependence of K_1 in nickel,

$$K_1(T) = K_{10}[1-1.74(T/T_C)](M/M_0)^{10}.$$

In anticipation of the result of this paper, that the pseudodipolar coupling gives rise to the tenth power law, he has suggested that the linear dependence on T which appears in this formula is due to the variation of

[14] F. Keffer, Phys. Rev. 100, 1692 (1955).
[15] T. Kasuya, J. Phys. Soc. Japan 11, 944 (1956). Because of a numerical error, the value of K_{10} in this paper is too small by a factor of 2. As a result his power law is too large by the factor 2.
[16] W. J. Carr, Jr., Phys. Rev. 109, 1971 (1958).

the coupling parameter with the temperature. It should be noted, however, that at very low temperatures data is scarce and that the empirical formula fits best at the higher temperatures, especially where K_1 becomes positive. It appears then that no clear-cut comparison of this theory with experiment may be made at this time.

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APPENDIX A

We evaluate the second order contribution to the energy. The part of Eq. (14) which is independent of the spin-wave occupation numbers, $E_2{}^0$, may be written

$$E_2{}^0 = -\frac{\epsilon^2}{4} \sum_{l_1,l_2} (l_1{}^+)^2 (l_2{}^-)^2 \sum_k \epsilon_k{}^{-1}$$
$$\times \exp[i\mathbf{k} \cdot (\mathbf{l}_1+\mathbf{l}_2)]. \quad (A1)$$

The sum on $\mathbf{k}$ is determined by the value of the summand for large $\mathbf{k}$ because of the density of states. In that region the spin-wave energy, ϵ_k, may be set equal to just the exchange part, and

$$E_2{}^0 = -\frac{\epsilon^2 N}{16\pi J} \sum_{l_1,l_2} (l_1{}^+)^2 (l_2{}^-)^2 b_{l_1+l_2}. \quad (A2)$$

The b_r were defined in Sec. V. A number of them were evaluated by Van Peipe.[4] We have calculated the others in which we are interested by use of Eq. (38) as well as by numerical integration. In our notation, b_{fgh} is that b_r associated with the lattice vector whose components along the cube edges are, in units of the length of a component of a nearest neighbor vector, the integers f, g, h. The values of interest are:

Simple cubic: $b_{000}=3.23$, $b_{100}=1.14$, $b_{200}=0.82$, $b_{110}=0.70$, $b_{111}=0.56$.

Body centered: $b_{000}=2.187$, $b_{111}=0.616$, $b_{200}=0.456$, $b_{220}=0.366$, $b_{222}=0.317$.

Face centered: $b_{000}=1.395$, $b_{110}=0.348$, $b_{200}=0.231$, $b_{220}=0.168$, $b_{211}=0.190$.

Because of the symmetry, the value of any b_{fgh} remains unchanged if its indices are permutted in any way or reversed in sign. At this point we are ready to sum over $\mathbf{l}_2$ in Eq. (A2). The technique used here will also be used in Appendix B to get the third order energy and in Sec. V of the paper in the determination of the ρ_1. Those terms for which $\mathbf{l}_2=\pm\mathbf{l}_1$ are separated from the rest. For the remaining terms, except in the case of the face-centered cubic, a common coefficient exists. One

may write

$$\sum_{l_2} (l_2{}^-)^2 b_{l_1+l_2} = (b_{l_1+l_1}+b_{l_1-l_1}-F)(l_1{}^-)^2 + F \sum_{l_2}(l_2{}^-)^2.$$

In the case of the face-centered cubic one must add the term

$$(2b_{200}-F)[(l_1{}^*)^-]^2,$$

where by $\mathbf{l}_1{}^*$ we mean either of the two nearest neighbor vectors of the face centered cubic which are normal to $\mathbf{l}_1$. For the simple cubic lattice, body centered and face centered cubics, respectively, $F=2b_{110}$, $b_{220}+b_{002}$, and $b_{211}+b_{011}$. Because of the cubic symmetry $\sum_{l_2}(l_2{}^-)^2=0$. The coefficient of $(l_1{}^-)^2$ shall be called $4\pi\nu_j$, and we have for the simple cubic, $4\pi\nu_j=b_{000}+b_{200}-2b_{110}=2.65$, for the body centered, $4\pi\nu_j=b_{000}+b_{222}-b_{220}-b_{002}=1.682$, for the face centered, $4\pi\nu_j=b_{000}+b_{220}-b_{221}-b_{011}=1.025$. The coefficient of $[(l_1{}^*)^-]^2$, the extra term for the face-centered cubic, is $B=-0.076$.

Equation (A2) may be simplified to

$$E_2{}^0 = -\frac{\epsilon^2 N}{4J}\left[\nu_j \sum_1 (l^+l^-)^2 + \frac{B}{4\pi}\sum_1(l^+l^{*-})^2\right], \quad (A3)$$

where B is defined only for the face-centered cubic. In the usual order, it may be shown that

$$\sum_1(l^+l^-)^2=4(1-\Gamma), \quad \frac{32}{9}(1+\Gamma), \quad 2(3+\Gamma), \quad (A4)$$

and for the face-centered cubic

$$\sum_1(l^+l^{*-})^2=2(1-3\Gamma). \quad (A5)$$

The last term in $E_2{}^n$ [Eq. (14)] has a finite value at $\mathbf{k}=0$, where $\mathbf{k}=\mathbf{k}_1+\mathbf{k}_2$. For small $\mathbf{k}$ this term is

$$-\frac{\epsilon^2}{JN} \sum_{l_1,l_2} l_1{}^+l_1{}^z l_2{}^- l_2{}^z \sum_k n_k$$
$$\times [1+\exp(-i\mathbf{k}\cdot\mathbf{l}_1)][1+\exp(-i\mathbf{k}\cdot\mathbf{l}_2)]$$
$$\times \exp[i(\mathbf{k}/2)\cdot(\mathbf{l}_1+\mathbf{l}_2)] \sum_{k_1} \frac{\exp[i\mathbf{k}_1\cdot(\mathbf{l}_1+\mathbf{l}_2)]}{\sum_1(1-\cos\mathbf{k}\cdot\mathbf{l})}.$$

The sum on $\mathbf{k}_1$ is related to the b_r and one can show this second order term in $\mathfrak{U}$ to be

$$-\sum_k n_k(4\epsilon^2/J)\left[\nu_j \sum_1(l^z)^2 l^+l^- + (B/4\pi)\sum_1 l^+l^z l^{*-}l^{*z}\right], \quad (A6)$$

except for terms in k^2. In the usual order again one finds

$$\sum_1(l^z)^2 l^+l^- = 4\Gamma, \quad \frac{16}{9}(1-2\Gamma), \quad 2(1-\Gamma), \quad (A7)$$

and for the face-centered cubic

$$\sum_1 l^+l^z l^{*-}l^{*z}=2(3\Gamma-1). \quad (A8)$$

Then, for the simple cubic, body-centered and face-

centered cubic lattices we have, respectively, neglecting terms dependent on k,

$$E_2{}^n = \nu_j N (\epsilon^2/J)(\Gamma-1) - 20\nu_j(\Gamma-\tfrac{1}{5})(\epsilon^2/J)\sum_k n_k,$$

$$E_2{}^n = -(8/9)\nu_j N(\epsilon^2/J)(\Gamma+1)$$
$$+(160/9)\nu_j(\Gamma-\tfrac{1}{5})(\epsilon^2/J)\sum_k n_k,$$

$$E_2{}^n = -N(\epsilon^2/J)[\tfrac{1}{2}\nu_j(3+\Gamma)+(B/8\pi)(1-3\Gamma)]$$
$$+(\epsilon^2/J)[10\nu_j(\Gamma-\tfrac{1}{5})+(5B/2\pi)(1-3\Gamma)]\sum_k n_k.$$

Notice that the coefficient of Γ in the energy per spin wave is exactly $20/N$ times the coefficient of Γ in the 'zero-point energy, independent of the evaluation of the parameters ν_j and B.

APPENDIX B

We calculate the third order contribution to the energy. If products of the n_k are neglected,

$$\langle n | \mathcal{U}a^{-1}\mathcal{K}'a^{-1}\mathcal{U} | n \rangle$$

$$= -\frac{\epsilon^2 J}{4N}\sum_{k,k'}\frac{v_k v_{k'}{}^*}{\epsilon_k\epsilon_{k'}}\sum_1 \exp(i\mathbf{k}'\cdot\mathbf{l})$$

$$\times(\cos\mathbf{k}\cdot\mathbf{l}-1)\left(1-\frac{4}{N}\sum_{k''}n_{k''}+2n_k+2n_{k'}\right). \quad (B1)$$

It may be shown that the effect of the B parameter is negligible beyond the second order as considered in Appendix A. By the methods of Appendix A then it may be shown that

$$\sum_{k'}\sum_1\frac{v_{k'}{}^*}{\epsilon_{k'}}\exp(i\mathbf{k}'\cdot\mathbf{l})(\cos\mathbf{k}\cdot\mathbf{l}-1)=\nu_j\frac{N}{J}v_k{}^*,$$

and

$$\sum_k\sum_1\frac{v_k}{\epsilon_k}\exp(i\mathbf{k}'\cdot\mathbf{l})(\cos\mathbf{k}\cdot\mathbf{l}-1)=\nu_j\frac{N}{J}v_{k'},$$

when substituted into Eq. (B1). It follows that

$$\langle n | \mathcal{U}a^{-1}\mathcal{K}'a^{-1}\mathcal{U} | n \rangle$$

$$= -\nu_j\left[\frac{\epsilon^2}{4}\sum_k\frac{|v_k|^2}{\epsilon_k}\left(1+4n_k-\frac{4}{N}\sum_{k'}n_{k'}\right)\right]. \quad (B2)$$

Again neglecting products of the n_k and for $\mathbf{k}+\mathbf{k}'$ small,

$$\langle n | \mathcal{U}a^{-1}\mathcal{K}'a^{-1}\mathcal{U} | n \rangle$$

$$= -\nu_j\frac{2\epsilon^2}{N}\sum_{k,k'}\frac{(u_k+u_{k'})(u_k{}^*+u_{k'}{}^*-u_{k+k'}{}^*)}{\epsilon_k+\epsilon_{k'}-\epsilon_{k+k'}}$$

$$\times n_{k+k'}. \quad (B3)$$

Except for the term containing $u_{k+k'}{}^*$ this is just $\nu_j\langle n | \mathcal{U}a^{-1}\mathcal{U} | n \rangle$. This other term vanishes for $\mathbf{k}+\mathbf{k}'=0$, and may be neglected otherwise since the coefficient is small compared to that of the term we keep.