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PHYSICAL REVIEW B

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# Ferromagnetic Stability and Elementary Excitations in a $S = \frac{1}{2}$ Heisenberg Ferromagnet with Dipolar Interaction by the Green's-Function Method

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The temperature dependence of the elementary excitation energy in a dipolar Hamiltonian has been studied by the Green's-function equation-of-motion method. The result differs from that given by Charap and reduces to it for B = F = 0. The magnetization calculated at zero order of an iterative procedure coincides with the Holstein-Primakoff result. The criterion of stability of the ferromagnetic state has been found to be temperature dependent.

#### I. INTRODUCTION

Several models have been introduced in order to explain the properties of 3d electrons in magnetic solids in connection with the concept of itinerant versus localized nature of these electrons. Considerable interest has also been shown in recent years by experimentalists on the reliability of these models. In particular an extensive effort has been made in two directions: measuring the magnon dispersion law for a wide range of momentum transfer and studying their temperature dependence.<sup>1</sup>

The dispersion law has been studied as a function of the temperature for various materials with different experimental methods such as spin-wave resonance, <sup>2</sup> neutron diffraction, energy analysis by triple-axis spectrometry, and small-angle scattering techniques.<sup>3</sup> Since the temperature dependence is different in different models, we can distinguish among them.

Theoretically the temperature dependence of the magnon dispersion law in the Heisenberg model has been found by a perturbation method<sup>4</sup> and by solving temperature-dependent Green's-function equations of motion in some approximation.<sup>5</sup>

In this paper we consider anisotropy effects. We assume a more general Hamiltonian as a sum of the

Heisenberg and dipolar terms:

$$H = H_e + H_d ,$$

where

$$H_{e} = -\sum_{im} J_{im} \vec{\mathbf{S}}_{i} \cdot \vec{\mathbf{S}}_{m} + 2\mu_{B} \Im C \sum_{i} S_{i}^{x} , \qquad (1)$$

$$H_{d} = \sum_{i > m} \frac{1}{2} d_{im} [\vec{\mathbf{S}}_{i} \cdot \vec{\mathbf{S}}_{m} - 3r_{im}^{2} (\vec{\mathbf{S}}_{i} \cdot \vec{\mathbf{r}}_{im}) (\vec{\mathbf{S}}_{m} \cdot \vec{\mathbf{r}}_{im})] .$$

Magnetic quadrupole interactions, also invoked to explain ferromagnetic anisotropy, are neglected; in fact their ratio to the dipole-dipole interaction need only be  $\sim \frac{1}{10}$ .<sup>6</sup> We can express  $H_d$  in terms of raising and lowering operators:

 $H_d = H_d^0 + H_d^* + H_d^- + H_d^{**} + H_d^{-*},$ 

where

$$H_{d}^{0} = \sum_{l \neq m} E_{lm} (\vec{S}_{l} \cdot \vec{S}_{m} - 3S_{l}^{z}S_{m}^{z})$$

$$H_{d}^{+} = \sum_{l \neq m} F_{lm}S_{l}^{+}S_{m}^{z},$$

$$H_{d}^{-} = \sum_{l \neq m} F_{lm}^{*}S_{l}^{-}S_{m}^{z},$$

$$H_{d}^{**} = \sum_{l \neq m} B_{lm} S_{l}^{*} S_{m}^{*} ,$$

$$H_{d}^{--} = \sum_{l \neq m} B_{lm}^{*} S_{l}^{-} S_{m}^{-} ,$$

$$2E_{lm} = -\frac{1}{4} d_{lm} [1 - 3(r_{lm}^{s} / r_{lm})^{2}] ,$$

$$2F_{lm} = -\frac{3}{2} d_{lm} [r_{lm}^{s} (r_{lm}^{x} - ir \frac{y}{lm}) / r_{lm}^{2}] ,$$

$$2B_{lm} = -\frac{3}{8} d_{lm} [(r_{lm}^{x} - ir \frac{y}{lm}) / r_{lm}]^{2} ,$$
(2)

with standard use of symbols.

If we retain only  $H_d^0$  in the dipolar Hamiltonian, the magnon energy presents, as has been shown by Charap, <sup>7</sup> a  $T^{3/2}$  term besides the  $T^{5/2}$  term appearing in the isotropic case.

In our work we also take into account  $H_d^{**}$  and  $H_d^{--}$  which play an important role in the Holstein-Primakoff (HP) theory of anisotropy.<sup>8</sup> These terms cannot be neglected with respect to  $H_d^0$ , since  $B_{im}$  and  $E_{im}$  are of the same order of magnitude and there is no *a priori* reason for assuming that  $S^*S^*$  (and  $S^-S^-$ ) terms do not contribute at low temperature. The unrenormalized spin-wave energy found by HP,<sup>8</sup> to which our calculation reduces for T=0, confirms our statement.

We shall express the Hamiltonain in terms of Pauli operators in the spin- $\frac{1}{2}$  case and write the Green's-function equations of motion for their transforms in wave-vector space. We break the chain of equations at first order. This procedure is justified by the results which we get in the Heisenbergmodel case and which can be compared with wellknown calculations.<sup>5</sup>

The dependence of magnon energy on temperature will be derived. Moreover it is well known that the dipolar Hamiltonian must satisfy a particular criterion for the stability of the ferromagnetic state. This criterion is temperature dependent and reduces

to that of 
$$HP^8$$
 at  $T=0$ .

# II. DISPERSION LAW: GREEN'S-FUNCTION EQUATION OF MOTION

For simplicity we shall limit ourselves to the case in which each atom has only a single ferromagnetic electron. Under this hypothesis we can introduce the Pauli operators

$$S_m^* = b_m$$
,  $S_m^- = b_m^*$ ,  $S_m^* = \frac{1}{2} - n_m$ ,  $n_m = b_m^* b_m$ , (3)

which satisfy the following commutation relations:

$$\begin{bmatrix} b_m, b_n^{\dagger} \end{bmatrix} = (1 - 2n_m)\delta_{mn}, \qquad \begin{bmatrix} b_m, b_n \end{bmatrix} = 0 ,$$
  
$$b_n^2 = (b_n^{\dagger})^2 = 0 .$$
(4)

The general Hamiltonian takes the form

$$H = C_{0} + \sum_{Im} (J_{Im} + 2E_{Im} + 2\mu_{B} \Im C) n_{I} - \sum_{Im} (J_{Im} - E_{Im}) b_{I}^{\dagger} b_{m}$$
$$+ \sum_{Im} (B_{Im} b_{I} b_{m} + B_{Im}^{*} b_{I}^{\dagger} b_{m}^{\dagger}) - \sum_{Im} (F_{Im} b_{I} n_{m} + F_{Im}^{*} b_{I}^{\dagger} n_{m})$$
$$- \sum_{Im} (J_{Im} + 2E_{Im}) n_{I} n_{m} .$$
(5)

Let us introduce the Fourier transforms of  $b_m$  and  $b_m^{\dagger}$ :

$$b_{k} = \frac{1}{\sqrt{N}} \sum_{m} e^{i\vec{\mathbf{k}}\cdot\vec{\mathbf{r}}} b_{m}, \quad b_{k}^{\dagger} = -\frac{1}{\sqrt{N}} \sum_{m} e^{-i\vec{\mathbf{k}}\cdot\vec{\mathbf{r}}} b_{m}^{\dagger}. \quad (6)$$

It is an easy matter to verify that the following commutation rules in the wave-vector space hold as a consequence of the commutation relations [Eq. (4)] in the direct space:

$$[b_k, b_{k'}] = 0 , \quad [b_k, b_{k'}^{\dagger}] = \delta_{kk'} - (2/N) \sum_m e^{i(\vec{k} - \vec{k'}) \cdot \vec{\pi}_m} n_m .$$
(7)

Moreover the summation rule  $\sum_k b_{\lambda-k} b_{k+\rho} = 0$  holds for every  $\lambda$  and  $\rho$  as a consequence of the condition  $b_m^2 = 0$ . We can rewrite the Hamiltonian in terms of these operators:

$$H = C_{0} + \sum_{k} \left\{ \left[ J(0) + 2E(0) \right] - \left[ J(k) - E(\hat{k}) \right] + 2\mu_{B}\mathcal{K} \right\} b_{k}^{\dagger} b_{k} + \left[ B(\hat{k})b_{k}b_{-k} + B^{*}(\hat{k})b_{k}^{\dagger}b_{-k}^{\dagger} \right] \\ - \frac{1}{\sqrt{N}} \sum_{kk'} \left\{ F\left( \frac{\vec{k} - \vec{k'}}{|\vec{k} - \vec{k'}|} \right) b_{k-k'} b_{k}^{\dagger} b_{k'} + F^{*}\left( \frac{\vec{k} - \vec{k'}}{|\vec{k} - \vec{k'}|} \right) b_{k}^{\dagger} b_{k'} b_{k-k'} \right\} - \frac{1}{N} \sum_{kk'\rho} \left[ J(\rho) + 2E(\hat{\rho}) \right] b_{k}^{\dagger} b_{k+\rho} b_{k'}^{\dagger} b_{k'-\rho} , \qquad (8)$$

where

$$J(k) = \sum_{m} J_{lm} e^{i\vec{\mathbf{x}}\cdot\vec{\mathbf{r}}_{lm}}, \quad E(\hat{k}) = \sum_{m} E_{lm} e^{i\vec{\mathbf{x}}\cdot\vec{\mathbf{r}}_{lm}},$$
$$F(\hat{k}) = \sum_{m} F_{lm} e^{i\vec{\mathbf{x}}\cdot\vec{\mathbf{r}}_{lm}}, \quad B(\hat{k}) = \sum_{m} B_{lm} e^{i\vec{\mathbf{x}}\cdot\vec{\mathbf{r}}_{lm}},$$

We find

$$2E(\hat{k}) = 4\pi M_0 \sin^2 \theta_k, \quad 2B(\hat{k}) = 4\pi M_0 \sin^2 \theta_k e^{-2i\phi_k}$$

in HP<sup>8</sup> notation. All these functions, except J(k), depend only on the direction of  $\vec{k}$ . Our  $B(\hat{k})$  and  $E(\hat{k})$  are one-half of those given by HP. The other coefficients relevant in our calculation are given by HP. The retarded Green's-function equation of motion for two operators A and B and a given Hamiltonian H in the  $\omega$  representation is in standard notation (see Ref. 9)

$$\omega \langle \langle A; B \rangle \rangle_{\omega} = (1/2\pi) \langle [A, B] \rangle + \langle \langle [A, H]; B \rangle \rangle_{\omega} .$$
(9)

Once the Green's function is known the mean value  $\langle BA \rangle$  can be written in terms of it:

$$\langle BA \rangle = \int_{-\infty}^{+\infty} d\omega \frac{\langle \langle A; B \rangle \rangle_{\omega + i\epsilon} - \langle \langle A; B \rangle \rangle_{\omega - i\epsilon}}{e^{\beta\omega} - 1} , (10)$$

where  $\beta = 1/k_B T$  in standard notation.

#### A. Heisenberg-Model Case

We first discuss the equations of motion for the isotropic exchange. We consider the retarded Green's function  $\langle \langle b_{\lambda^*} b_{\lambda^*}^{\dagger} \rangle \rangle$  and write for it the equation of motion assuming the Heisenberg Hamiltonian (with zero magnetic field):

$$\omega \langle \langle b_{\lambda}; b_{\lambda'}^{\dagger} \rangle \rangle = (1/2\pi) (1-2n) \delta_{\lambda\lambda'} + [J(0) - J(\lambda)] \langle \langle b_{\lambda}; b_{\lambda'}^{\dagger} \rangle \rangle - (2/N) \times \sum_{kk'} [J(\vec{k} - \vec{k}') - J(k')] \langle \langle b_{k}^{\dagger} b_{k'} b_{\lambda+k-k'}; b_{\lambda'}^{\dagger} \rangle \rangle .$$
(11)

This equation involves higher-order Green's functions. We expect that as a first approximation only those Green's functions of the general form  $\langle \langle b_k^{\dagger} b_k, b_x; b_{\lambda'}^{\dagger} \rangle \rangle$  will appreciably contribute to the sum for which at least one of the two indexes k' and x is equal to k, and we substitute  $b_k^{\dagger} b_k$  with its mean value  $\langle n_k \rangle$ . The equation becomes

$$\left\{\omega - \left[J(0) - J(\lambda) + (2/N)\sum_{k} \langle n_{k} \rangle \langle J(\lambda) + J(k) - J(k-\overline{\lambda}) - J(0) \rangle \right]\right\} \langle \langle b_{\lambda}; b_{\lambda'}^{\dagger} \rangle = \left[(1 - 2n)2\pi\right] \delta_{\lambda\lambda'} .$$
(12)

The pole of the Green's function gives the excitation energy which is formally identical to the formula given by Brout and Englert<sup>10</sup> without proof and by Tahir-Kheli and ter Haar<sup>5</sup> in the first part of their second paper on the subject, on the basis of a firstorder Green's-function calculation.

By using formulas (10) and (12) we get for  $\langle n_k \rangle$  the following equation:

We get the following equations:

$$\langle n_k \rangle = (1 - 2n) [e^{\beta \omega_k} - 1]^{-1},$$
 (13)

which differs from the Tahir-Kheli-ter Haar formula<sup>5</sup> for the presence of a factor 1 - 2n instead of 1. The extra factor has its origin in the fact that Pauli operators are not bosonlike as are Dyson operators used by Tahir-Kheli and ter Haar.<sup>5</sup> They can be considered as approximately bosons only in the limit of low temperature when 2n is negligible compared to 1.

Equation (13) is an integral equation because its second side is a functional of  $\langle n_k \rangle$  through n and  $\omega_k$ . We can solve it by an iterative procedure. At zero order we put n = 0 and  $\langle n_k \rangle = 0$  on the right-hand side of the equation. This is correct for low temperatures. In this limit our results coincide with those of previous authors and give dependence  $T^{5/2}$  for the energy and  $T^{3/2}$  and  $T^{5/2}$  for the magnetization.

So we think this procedure will give a simple and essentially correct method for investigating the anisotropy effects at least in the limit of low temperatures.

#### **B.** Anisotropic Effects

We take into account  $H_d^{**}$  and  $H_d^{-*}$  terms but for simplicity we neglect in the full Hamiltonian the terms  $H_d^*$  and  $H_d^-$ , the contribution of which at low temperature will be small compared to that of  $H_d^{**}$ and  $H_d^{-*}$ 

$$\sum_{lm} F_{lm} b_l n_m \sim \sum_{T/T_c \ll 1} \sum_{lm} F_{lm} b_l \langle n \rangle = 0 \text{ as } \sum_m F_{lm} = 0$$

in an infinite lattice) even if the moduli of the coefficients F and B are of the same order of magnitude. The terms  $H_d^{++}$  and  $H_d^{--}$  cannot be discarded with respect to  $H_d^0$  because  $|B_{Im}|$  and  $E_{Im}$  are of the same order of magnitude, as can be seen in the definition (2). In the first-order decoupling we need only two Green's functions  $\langle\langle b_{\lambda}; b_{\lambda'}^{\dagger} \rangle\rangle$  and  $\langle\langle b_{-\lambda}^{\dagger}; b_{\lambda'}^{\dagger} \rangle\rangle$ ; hence we must calculate only the commutator  $[b_{\lambda}, H]$  since  $[b_{\lambda}^{\dagger}, H] = -[b_{\lambda}, H]^{\dagger}$  as a consequence of a Hermitian property of the Hamiltonian.

$$\omega \langle \langle b_{\lambda}; b_{\lambda'}^{\dagger} \rangle \rangle = \frac{1-2n}{2\pi} \delta_{\lambda\lambda'} + A(\vec{\lambda}) \langle \langle b_{\lambda}; b_{\lambda'}^{\dagger} \rangle \rangle - \frac{2}{N} \sum_{kk'} \left[ \overline{A}(\vec{k}) + \overline{J}(\vec{k} - \vec{k}') \right]$$

$$\times \langle \langle b_{k}^{\dagger} b_{k'} b_{k+\lambda-k'}; b_{\lambda'}^{\dagger} \rangle \rangle + 2B^{*}(\hat{\lambda}) \langle \langle b_{-\lambda}^{\dagger}; b_{\lambda'}^{\dagger} \rangle \rangle - \frac{2}{N} \sum_{kk'} B^{*}(\hat{k}) \langle \langle b_{k'}^{\dagger} b_{-k}^{\dagger} b_{k'+\lambda-k} + b_{k'}^{\dagger} b_{k}^{\dagger} b_{k+\lambda-k'}; b_{\lambda'}^{\dagger} \rangle \rangle ,$$

$$\omega \langle \langle b_{-\lambda}^{\dagger}; b_{\lambda'}^{\dagger} \rangle \rangle = -A(\vec{\lambda}) \langle \langle b_{-\lambda}^{\dagger}; b_{\lambda'}^{\dagger} \rangle \rangle + \frac{2}{N} \sum_{kk'} \left[ \overline{A}(\vec{k}) + \overline{J}(\vec{k} - \vec{k}') \right]$$

$$\times \langle \langle b_{k'}^{\dagger} b_{k-\lambda-k'}^{\dagger} b_{k}; b_{\lambda'}^{\dagger} \rangle \rangle - 2B(\hat{\lambda}) \langle \langle b_{\lambda}; b_{\lambda'}^{\dagger} \rangle \rangle + \frac{2}{N} \sum_{kk'} B(\hat{k}) \langle \langle b_{k'-\lambda-k}^{\dagger} b_{k} b_{k'} + b_{k+\lambda+k'}^{\dagger} b_{k} b_{k'}; b_{\lambda'}^{\dagger} \rangle \rangle ,$$

$$(14)$$

2245

where  $A(\overline{\lambda})$ ,  $\overline{A}(\overline{\lambda})$ , and  $\overline{J}(\overline{\lambda})$  are

$$\begin{split} A(\lambda) &= J(0) + 2E(0) - J(\lambda) - E(\hat{\lambda}) + 2\mu_B \mathcal{K} ,\\ \overline{A}(\hat{\lambda}) &= E(\hat{\lambda}) - J(\lambda) ,\\ \overline{J}(\hat{\lambda}) &= J(\lambda) + 2E(\hat{\lambda}) . \end{split}$$

Now we proceed in exactly the same way as for the Heisenberg case in order to decouple the higherorder Green's functions: We take only the diagonal terms in the sums and substitute for  $n_k$  its mean value  $\langle n_k \rangle$ :

$$\begin{split} [\omega - W_0(\hat{\lambda})] \langle \langle b_{\lambda}; b_{\lambda}^{\dagger}, \rangle \rangle &- \{B^*(\hat{\lambda}) - (2/N) \\ \times \sum_k [B^*(\hat{\lambda}) + B^*(\hat{k})] \langle n_k \rangle \} \langle \langle b_{-\lambda}^{\dagger}; b_{\lambda'}^{\dagger}, \rangle \rangle \\ &= [(1 - 2n)/2\pi] \delta_{\lambda\lambda'}, \quad (15) \end{split}$$

$$\begin{split} \left\{ B(\hat{\lambda}) - (2/N) \sum_{k} \left[ B(\hat{\lambda}) + B(\hat{k}) \right] \langle n_{k} \rangle \right\} \\ & \times \langle \langle b_{\lambda}; b_{\lambda'}^{\dagger} \rangle \rangle + \left[ \omega + W_{0}(\hat{\lambda}) \right] \langle \langle b_{-\lambda}^{\dagger}; b_{\lambda'}^{\dagger} \rangle \rangle = 0 , \end{split}$$

where

$$W_{0}(\vec{\lambda}) = A(\vec{\lambda}) + \frac{2}{N} \sum_{k} \left\{ [J(\lambda) + J(k) - J(k - \lambda) - J(0)] - \left[ 2E\left(\frac{\vec{k} - \vec{\lambda}}{|\vec{k} - \vec{\lambda}|}\right) + 2E(0) + E(\hat{k}) + E(\hat{\lambda}) \right] \right\} \langle n_{k} \rangle .$$

It is easy to verify that the poles of the Green's functions are the solution of a second-order equation:

$$\omega = \pm \left\{ \left[ W_0(\vec{\lambda}) \right]^2 - 4 \left| B(\hat{\lambda}) - (2/N) \right. \right. \\ \left. \times \sum_k \left[ B(\hat{\lambda}) + B(\hat{k}) \right] \left< n_k \right> \left|^2 \right>^{1/2} = \pm W(\vec{\lambda}) \right.$$
(16)

Of course one must have

$$|W_{0}(\hat{\lambda})|^{2} > 4|B(\hat{\lambda}) - (2/N)\sum_{k} [B(\hat{\lambda}) + B(\hat{k})]\langle n_{k}\rangle|^{2}$$
(16')

for a stable ferromagnetic state (otherwise  $\omega$  would be pure imaginary).

The given criterion for the stability of the ferromagnetic state is temperature dependent through  $\langle n_b \rangle$ . The Green's function  $\langle \langle b_\lambda; b_\lambda^{\dagger} \rangle \rangle$  is given by

$$\langle\langle b_{\lambda}; b_{\lambda}^{\dagger} \rangle\rangle = (1 - 2n) \frac{\omega + W_0(\tilde{\lambda})}{\omega^2 - [W(\tilde{\lambda})]^2} .$$
 (17)

Of course,

$$\langle \langle b_{\lambda}; b_{\lambda}^{\dagger} \rangle \rangle_{\omega + i\epsilon} - \langle \langle b_{\lambda}; b_{\lambda}^{\dagger} \rangle \rangle_{\omega - i\epsilon} = -2\pi i (1 - 2n) \left[ \omega + W_0(\bar{\lambda}) \right]$$
$$\times \delta(\omega^2 - \left[ W(\bar{\lambda}) \right]^2) . \quad (18)$$

By using the identity

$$\delta(\omega^2 - [W(\vec{\lambda})]^2) = (1/2\omega) [\delta(\omega - W(\vec{\lambda})) + \delta(\omega + W(\vec{\lambda}))]$$

and inserting the formula in the spectral representation (10), we have

$$\langle n_{\lambda} \rangle = (1 - 2n) \left[ \frac{1}{2} \left( \frac{1}{e^{\beta W} - 1} + \frac{1}{e^{-\beta W} - 1} \right) + \frac{W_0}{2W} \left( \frac{1}{e^{\beta W} - 1} - \frac{1}{e^{-\beta W} - 1} \right) \right].$$
 (19)

This equation can be solved with an iterative procedure. At zero order we must put n = 0 and  $\langle n_k \rangle = 0$  in the right-hand side of the equation. It is equivalent to the following substitutions:  $W_0(\bar{\lambda}) \rightarrow A(\bar{\lambda})$  and  $W(\bar{\lambda}) \rightarrow \{[A(\bar{\lambda})]^2 - 4 | B(\hat{\lambda})|^2\}^{1/2}$ . For the Heisenberg model the zero-order magnon number  $\langle n_{\lambda}^{0} \rangle$  will be

$$\langle n_{\lambda}^{0} \rangle = [e^{\beta [J(0) - J(\lambda)]} - 1]^{-1}$$
 (20)

Because the anisotropic effects are quantitatively small, the difference  $\langle n_{\lambda} \rangle - \langle n_{\lambda}^{0} \rangle$  will also be small. So we can use the trivial identity  $\langle n_{\lambda} \rangle = (\langle n_{\lambda} \rangle - \langle n_{\lambda}^{0} \rangle) + \langle n_{\lambda}^{0} \rangle$  and substitute in the difference, for all the integrals we have to calculate, the exponential with their first-order approximation. In this way we obtain

$$\langle n_{\lambda} \rangle \rightarrow k_{B}T \left( \frac{A(\overline{\lambda})}{[A(\overline{\lambda})]^{2} - 4 |B(\overline{\lambda})|^{2}} - \frac{1}{J(0) - J(\lambda)} \right) + \frac{1}{e^{B[J(0) - J(\lambda)]} - 1} .$$
(21)

In the exponential we take the first- and when necessary the second-order expansion of  $J(0) - J(\lambda)$ , while in the other terms [including  $A(\overline{\lambda})$ ] it is sufficient to take the first-order expansion  $J(0) - J(\lambda)$  $\simeq J a^2 \lambda^2$ . We get the same magnetization found by HP.<sup>8</sup> This can be understood if we consider that the T = 0 spin-wave energy determines the lowtemperature dependence of the magnetization and our renormalized spin-wave energy reduces to that of HP in this (T = 0) limit.

We discuss now  $W_0(\vec{\lambda}, T)$  which we will derive in the Appendix:

$$W_{0}(\vec{\lambda}, T) - A(\vec{\lambda}) = J a^{2} \lambda^{2} (\alpha_{1} T - \alpha_{5/2} T^{5/2})$$
  
- 3 [E(\lambda) + \int d \hat{k} E(\hat{k})] (\gamma\_{1} T - \alpha\_{3/2} T^{3/2})  
- (2/N) \sum\_{\vec{r}} E(\hat{\gamma}) r^{2} (1 - e^{i\lambda \cdot \vec{r}}) (\alpha\_{1} T - \alpha\_{5/2} T^{5/2}) .  
(22)

We get similar results for

$$\sum_{k} \left[ B(\hat{\lambda}) + B(\hat{k}) \right] \langle n_{k} \rangle = \left[ B(\hat{\lambda}) + \int d\hat{k} B(\hat{k}) \right]$$
$$\times \left( \gamma_{1} T - \alpha_{3/2} T^{3/2} \right) . \tag{23}$$

A linear T term appears with opposite sign from the other terms of the temperature series. We want to compare the linear term in respect to the  $T^{3/2}$  term. If  $\gamma_1 T \sim \alpha_{3/2} T^{3/2}$  their effect cancels out. If we choose a temperature range in which  $\gamma_1 T \sim \frac{1}{2} \alpha_{3/2} T^{3/2}$  it is possible to appreciate the deviation of the temperature law from the  $T^{3/2}$  behavior. It is impossible to find a temperature range in which  $\gamma_1 T$  overcomes the other term. By using the HP expression for  $\gamma_1$  we find that the condition  $\gamma_1 T / \alpha_{3/2} T^{3/2} \sim \frac{1}{2}$  is satisfied for nickel and iron at  $T \sim 60$  °K and H about three or four times  $4\pi M_0$  in HP notation. The situation is more favorable in the hcp cobalt, the anisotropy of which is much stronger. In this case, however, the calculation cannot be directly applied since we assumed in our treatment the cubic symmetry.

Since the magnon energy is given by formula (16), we see that the dependence on temperature is very cumbersome. However, if we develop  $\omega_{\lambda}(T)$  as a function of T we find that at the very low temperature in which we are interested, it is still linear in T. In inelastic neutron scattering only a  $T^{5/2}$ term has so far been measured. In fact, its coefficient is much larger than the others appearing in formulas (22) and (23). We would expect, however, that the T and  $T^{3/2}$  terms can be observed only by looking at the directional dependence of the dispersion law and at relatively low temperature and with a magnetic field three or four times  $4\pi M_0$ .

### **III. CONCLUSIONS**

The Green's-function method allows us to find a temperature dependence in the dispersion law and in the criterion for the stability of the ferromagnetic state in the dipolar Hamiltonian. The method of decoupling the chain of equations in the first approximation is justified by the fact that the results are correct for the Heisenberg model at low temperatures (up to and including the  $T^{5/2}$  term in both the magnon energy and the magnetization). The divergence with other theories we get in the Heisenberg case for terms with power higher than  $\frac{5}{2}$  is due to the kinematical interaction arising from the nature of Pauli operators which only in the limit of low temperature can be considered boson-like operators.

The principal results we get are the following:

- Our magnetization coincides with that given by Holstein and Primakoff<sup>8</sup> and differs from what we could obtain from Charap's Hamiltonian.
- (ii) The temperature dependence of magnon energy differs from that given by Charap<sup>7</sup> as it should, because we retain more terms in the Hamiltonian. These terms cannot be neglected since the coefficients are of the same order of magnitude of those retained

by Charap. Moreover, if we also neglect the *B* terms (but not the *E* terms), the occupation number  $n_{\lambda}$  will still retain the form of Eq. (21) which simply follows from the statement  $\langle n_{\lambda} \rangle = \langle \langle n_{\lambda} \rangle - \langle n_{\lambda}^{0} \rangle \rangle + \langle n_{\lambda}^{0} \rangle$ ,  $\langle n_{\lambda}^{0} \rangle$  being the isotropic Heisenberg-model occupation number. And this implies a linear dependence of the magnon energy on the temperature.

(iii) The criterion of stability of the ferromagnetic state, which has been found temperature independent in the Holstein-Primakoff approximation,<sup>8</sup> really depends on temperature.

In our decoupling we used the simple Hartree-Fock approximation in which the correlation functions  $\langle b_k b_{-k} \rangle$  and  $\langle b_{-k}^{\dagger} b_k^{\dagger} \rangle$  are neglected. They are identically zero for B=0 and smaller than  $\langle b_k^{\dagger} b_k \rangle$ which we retained. However, they do not modify the behavior as a function of the temperature.

## APPENDIX

We wish to show how the temperature dependence of the magnon energy can be derived. We limit ourselves to the calculation of  $W_0(\vec{\lambda}, T)$ . The term  $\sum_k [B(\hat{\lambda}) + B(\hat{k})] \langle n_k \rangle$  can be deduced in a similar way.

We use the expression (15) for  $W_0(\vec{\lambda})$  and the approximation (21) for  $\langle n_k \rangle$ . Some coefficients in the Hamiltonian depend only on the direction of the wave vector; this is specified with the use of a caret on the wave vector:

$$W_{0}(\vec{\lambda}, T) - A(\vec{\lambda}) = \frac{2}{N} \sum_{k} \left\{ [J(\lambda) + J(k) - J(\vec{k} - \vec{\lambda}) - J(0)] - \left[ 2E\left(\frac{\vec{k} - \vec{\lambda}}{|\vec{k} - \vec{\lambda}|}\right) + 2E(0) + E(\hat{k}) + E(\hat{\lambda}) \right] \right\} \langle n_{k} \rangle .$$
(A1)

We consider first

$$(2/N) \sum_{k} \left[ J(\lambda) + J(k) - J(k - \lambda) - J(0) \right] \langle n_{k} \rangle$$
  
=  $(2/N) \sum_{\vec{\tau}} J(r) (1 - e^{i\vec{\lambda} \cdot \vec{\tau}}) \sum_{k} (e^{-i\vec{k} \cdot \vec{\tau}} - 1) \langle n_{k} \rangle,$   
(A2)

where use has been made of the definition  $J(q) = \sum_{r} J(r) e^{i\vec{q} \cdot \vec{r}}$  and the property J(r) = J(-r) which implies J(q) = J(-q). With the aid of Eq. (21), Eq. (A2) can be rewritten as

$$\frac{2}{N} \sum_{r} J(r) \left(1 - e^{i\vec{k} \cdot \vec{r}}\right) \sum_{k} \left(1 - e^{i\vec{k} \cdot \vec{r}}\right) \times \left[k_{B}T \left(\frac{A(\vec{k})}{[A(\vec{k})]^{2} - 4 |B(\vec{k})|^{2}} - \frac{1}{Ja^{2}k^{2}}\right) + \langle n_{k}^{0} \rangle\right],$$
(A3)

where the first term gives the linear contribution in *T* whereas the second one gives higher-order contribution in *T*. As far as the first term is concerned a good estimation of the coefficient of the linear term is obtained by retaining only terms of the order  $k^2$  in  $1 - e^{-i\vec{k}\cdot\vec{T}}$  following a similar argument in HP.<sup>8</sup> Similarly we evaluate the second term where the presence of the Bose occupation number  $\langle n_k^0 \rangle$  legitimates the expansion in  $k^2$  of the integrand at low temperatures. The second term can be written

$$\frac{2}{N} \sum_{k} \left\{ 2 \left[ E(\hat{\lambda}) + E(\hat{k}) + E\left(\frac{\vec{k} - \vec{\lambda}}{|\vec{k} - \vec{\lambda}|}\right) - E(0) \right] - 3 \left[ E(\vec{k}) + E(\vec{\lambda}) \right] \right\} \langle n_{k} \rangle \quad . \tag{A4}$$

Here the first contribution is

$$-\frac{2}{N}\sum_{\vec{r}} E(\hat{r}) (1 - e^{i\vec{\lambda} \cdot \vec{r}}) \frac{V}{(2\pi)^3} \int (\vec{k} \cdot \vec{r})^2 \\ \times \left\{ \left[ \frac{A(\vec{k})}{[A(\vec{k})]^2 - 4 |B(\hat{k})|^2} - \frac{1}{Ja^2k^2} \right] k_B T \\ + \frac{1}{e^{\beta Ja^2k^2} - 1} \right\} d\vec{k} .$$
 (A5)

The difference in the two cases is of course that J(r) is isotropic while  $E(\hat{r})$  is not.

The second contribution is

$$\frac{3}{N} \sum_{k} \left[ E(\hat{k}) + E(\hat{\lambda}) \right] \left\{ k_{B}T \left[ \frac{A(\vec{k})}{\left[ A(\vec{k}) \right]^{2} - 4 |B(\hat{k})|^{2}} - \frac{1}{Ja^{2}k^{2}} \right] + \frac{1}{e^{\beta Ja^{2}k^{2}} - 1} \right\}.$$
 (A6)

The integrals

$$\begin{aligned} &\alpha_{3/2} \ T^{3/2} = \int \frac{1}{e^{\beta J a^{2} k^{2}} - 1} \ d\vec{k} \ , \\ &\alpha_{5/2} \ T^{5/2} = \int \ \frac{(\vec{k} \cdot \hat{\gamma})^{2}}{e^{\beta J a^{2} k^{2}} - 1} \ d\vec{k} \ , \end{aligned}$$

<sup>1</sup>G. Shirane, V. J. Minkiewiez, and R. Nathans, J. Appl. Phys. <u>39</u>, 383 (1968).

<sup>2</sup>T. G. Phillips, Phys. Letters <u>17</u>, 11 (1965); R. Weber and P. E. Tannenwald, J. Phys. Chem. Solids <u>24</u>, 1357 (1963).

<sup>3</sup>M. W. Stringfellow, Proc. Phys. Soc. <u>1</u>, 950 (1968).
 <sup>4</sup>W. Marshall (unpublished); P. E. Tannenwald, J.
 Phys. Soc. Japan Suppl. <u>17</u>, 592 (1962); T. Oguchi,

Phys. Rev. <u>117</u>, 117 (1960).

<sup>5</sup>R. A. Tahir-Kheli and D. ter Haar, Phys. Rev. <u>127</u>, 95 (1962); J. F. Cooke and H. A. Gersch, *ibid*. <u>153</u>,

and

$$\int E(\hat{k}) \frac{1}{e^{\beta J a^2 k^2} - 1} d\vec{k}$$
$$= \int E(\hat{k}) d\hat{k} \int \frac{1}{e^{\beta J a^2 k^2} - 1} k^2 dk$$

can be evaluated in the standard way. The integral

$$\gamma_1 = \frac{V}{(2\pi)^3} \int \left[ \frac{A(\vec{k})}{[A(\vec{k})]^2 - 4 |B(\hat{k})|^2} - \frac{1}{Ja^2k^2} \right] d\vec{k}$$

has been evaluated by HP. We do not attempt to evaluate

$$\alpha_{1} = \frac{V}{(2\pi)^{3}} \int (\vec{k} \cdot \hat{r})^{2} \left[ \frac{A(\vec{k})}{[A(\vec{k})]^{2} - 4 |B(\hat{k})|^{2}} - \frac{1}{Ja^{2}k^{2}} \right] d\vec{k}$$

but we merely state that  $\alpha_1 \ll \gamma_1$ . In fact, the integrals can be written in polar coordinates:

$$\int k^4 dk \int (\hat{k} \cdot \hat{r})^2 f(\vec{k}) d\hat{k} \text{ and } \int k^2 dk \int f(\vec{k}) d\hat{k}$$

Both integrals get the main contribution from the region where

$$k^2 \sim \frac{4\pi \,\mu_{\beta} \,M_0 + 2\,\mu_{\beta}\,3\mathcal{C}}{J} \sim 10^{-3}$$

as has been stated by HP. It is obvious that since in this region  $k^2 \ll 1$  the integral involving the higher power of k is much less than the other:

 $\alpha_1 \sim 10^{-3} \gamma_1$ .

We can conclude

$$\begin{split} W_{0}(\vec{\lambda}, T) - A(\vec{\lambda}) &= Ja^{2}\lambda^{2} \left(\alpha_{1}T - \alpha_{5/2}T^{5/2}\right) \\ &- 3[E(\hat{\lambda}) + \int d\hat{k} \ E(\hat{k})] \ (\gamma_{1}T - \alpha_{3/2}T^{3/2}) \\ &- (2/N) \sum_{\vec{r}} \ E(\hat{r}) \ r^{2} \ (1 - e^{i\vec{\lambda} \cdot \vec{r}}) \ (\alpha_{1}T - \alpha_{5/2}T^{5/2}) \ , \end{split}$$

$$(A7)$$

where all the  $\alpha$  and  $\gamma$  are positive.

<sup>8</sup>T. Holstein and H. Primakoff, Phys. Rev. <u>58</u>, 1098 (1940).

<sup>9</sup>D. N. Zubarev, Usp. Fiz. Nauk <u>71</u>, 71 (1960) [Sov. Phys. Usp. <u>3</u>, 320 (1960)].

<sup>10</sup>R. Brout and F. Englert, Bull. Am. Phys. Soc. <u>6</u>, 55 (1961).

<sup>641 (1967);</sup> W. Marshall and G. Murray, J. Appl. Phys. <u>39</u>, 380 (1968).

<sup>&</sup>lt;sup>6</sup>F. Keffer, in *Handbuch der Physik*, edited by S. Flügge (Springer, Berlin, 1966), Vol. XVIII/2, Chap. III.

<sup>&</sup>lt;sup>7</sup>S. H. Charap, Phys. Rev. <u>136</u>, A1131 (1964).