Phys. Rev. 182, 1165 (1969).

[17] J. C. Love, F. E. Obenshain, and G. Czjzek, Phys. Rev. B 3, 2827 (1971).

[18] G. Fischer, A. Herr, and A. J. P. Meyer, J. Appl. Phys. 39, 545 (1968).

[19] J. W. Burton, in Oak Ridge National Laboratory Report N[illegible] ORNL-4743 (unpublished).

[20] H. H[illegible]. Wegener and F. E. Obenshain, Z. Physik 163, 17 [illegible]1).

[21] D. A. Shirley, S. S. Rosenblum, and E. Matthias, Phys. Rev. 170, 363 (1968).

[22] C. Kittel, *Introduction to Solid State Physics* (Wiley, New York, 1967), p. 435.

[23] A. A. Maradudin, in *Solid State Physics*, Vol. 18, edited by F. Seitz and D. Turnbull (Academic, New York, 1966), p. 389.

[24] A. J. F. Boyle and H. E. Hall, Rept. Progr. Phys. 25, 441 (1962).

[25] H. H. F. Wegener, *Der Mössbauer Effekt* (Bibliographisches Institut, Mannheim, 1965), pp. 61 and 109.

[26] R. M. Housley and F. Hess, Phys. Rev. 146, 517 (1966).

[27] A. P. Miller and B. N. Brockhouse, Can. J. Phys. 49, 704 (1971); and A. P. Miller (private communication).

[28] B. Mozer, K. Otnes, and V. W. Myers, Phys. Rev. Letters 8, 278 (1962).

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Spatially Random Heisenberg Spins at Very Low Temperatures. I. Dilute Ferromagnet*

Raza A. Tahir-Kheli
Department of Physics, Temple University, Philadelphia, Pennsylvania 19122

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A formulation is given for calculating magnetic-single-site density of states for a completely random dilute Heisenberg ferromagnet, with isotropic nearest-neighbor exchange, in the limit of very low temperatures. In conformity with Kohn's suggestion that the dynamics of sufficiently random many-body systems may be approximated by that of typical small neighborhoods, a consistent hierarchy of truncation schemes for the spatial matrix elements of the T matrix is described. The case of a drastically truncated Kohn neighborhood, consisting only of two neighboring sites, is worked out in detail. It is shown that for lattices without nearest-neighbor triangles, the given density of states exactly preserves the first four frequency moments. Moreover, for Bravais lattices with z nearest neighbors, all frequency moments of the density of states are given exactly to the two leading orders in z^{-1}. By analyzing the renormalization of the $K\to 0$ spin-wave energy, estimates for the critical temperature are obtained. In the present approximation, the magnetic long-range order cannot occur for magnetic concentrations which are $\leq 2/z$. For the simple-cubic lattice, numerical computations of the magnetic-single-site density of states and the real and imaginary parts of the coherent exchange are given for several concentrations.

I. INTRODUCTION

The ground state of a dense Heisenberg ferromagnet is exactly known. However, as soon as finite concentrations of nonmagnetic impurities are introduced, the system becomes a random coupled many-body system which cannot be solved exactly in arbitrary dimensionality. Brout[1] seems to have been the first one to seriously address himself to the question of the behavior of such a dilute Heisenberg ferromagnet as a function of the dilution. His analysis was rather formal, and although no precise results were recorded, a qualitative picture of the dependence of the Curie temperature $T_C(m)$ as a function of the magnetic concentration m was predicted. In the limit that the exchange interactions are extremely long ranged, the dependence of $T_C(m)$ on m was conjectured to be linear. For finite-range interactions, the linear dependence was conjectured to be confined to the concentrated region, while in the vicinity of a certain nonzero critical concentration m_c [m_c is the highest relative concentration of magnetic ions for which magnetic long-range order (LRO) does not occur] the behavior was expected to be more complicated.

The problem was later studied by Elliott[2] and Smart.[3] Elliott[2] used the constant-coupling two-particle cluster approximation of Kasteleijn and van Kranendonk.[4] For spin S and coordination number z, he estimated the critical concentration m_c as

$$m_c = (S+1)/S(z-1) \; . \qquad (1.1)$$

Smart[3] generalized the Bethe-Peierls-Weiss method for application to the classical spin[5] case (i.e., $S\to\infty$) and found the same result.

Charap[6] argued that because of the neglect of concentration fluctuations in the environment of the nearest-neighbor shell, the physics of the problem had been inadequately represented in this[3] treatment. Assuming that any magnetic atom i in the first shell is also subject to a fluctuating molecular field, being proportional to the number of its magnetic neighbors z_i in the given configuration, Charap[6] estimated that for nearest-neighbor isotropic Heisenberg exchange interactions, critical magnetic concentrations were 0.33 and 0.24, respectively, for the (simple cubic) sc and the bcc lattices.

Elliott[7] and Heap[7,8] studied the behavior of the paramagnetic susceptibility χ of the system as a power series in the concentration m. In two such analyses, critical temperatures were estimated by fixing the temperature T and finding the concentration $m(T)$ for which the susceptibility diverged. The results of these analyses confirmed Brout's conjecture: For concentrations near the dense limit, the results for T_C were largely linear but the extrapolation to the critical concentration limit was susceptible to large errors.

Perhaps, mention should also be made of the Elliott[9]-Rushbrooke[10] assertion that the magnitude of the "critical concentration is a topological property of the lattice and is the same in the Heisenberg and the Ising models and is independent of the spin S." Even though there are good physical reasons to be skeptical of this assertion,[8,11,12] the Ising model[13,14] and the related studies on the cluster size distributions in the percolation theory of the site problem,[15,16] predict the magnitude of the results in three dimensions to be roughly the same as those given by Charap.[6]

Excepting the work of Murray,[12] all the foregoing works[1–11,13–16] have made use of static formalisms. In other words, these works have not dealt with the dynamics of the problem. Murray's work attempts to compute the spin-wave energy $E_{\vec{K}}(m)$ as a function of the concentration m in the limit of long wavelengths, i.e., $\vec{K} \to 0$. Because magnetic LRO implies the existence of these spin waves, Murray has computed the concentration dependence of the transition temperature by equating $T_C(m)/T_C(1)$ to the corresponding ratio of the spin-wave energies $E_{\vec{K}}(m)/E_{\vec{K}}(1)$. This treatment has predicted results for m_c which are much higher than those given by the earlier theories.[1–10,13–16] However, because the validity of the variational principle used by Murray is not entirely beyond doubt,[17] the actual magnitude of the discrepancy between the earlier results for m_c and those given by Murray cannot be given too much importance.

Recently, two dynamical theories of the dilute ferromagnet have appeared in the literature.[18] Based on the cumulant decoupling-diagram method of Matsubara *et al.*,[19] expressions for the K-dependent equilibrium Green's function were obtained. Following Murray,[12] the renormalization of the long-wavelength spin wave was used to predict the concentration dependence of the Curie temperature $T_C(m)$.

These careful works have computed the dependence of $T_C(m)$ on m by using a series expansion for $E_{\vec{K}}(m)$ in powers of $(1-m)$. Computing the first two leading terms in this series exactly and the third term, i.e., $O(1-m)^2$ approximately, they have predicted a largely linear dependence of $T_C(m)$ on m. Extrapolating their results down to the critical region, they have also estimated the magnitude of the critical concentration m_c. For the sc lattice, their estimates for m_c, i.e., 0.329 (Kaneyoshi) and 0.362 (Edwards and Jones), seem to lie close to the results of the various static theories.[1–10,13–16]

In the present work we use a different approach to the solution of the dynamical problem. By interpreting the ideas of Kohn[20] rather loosely, we formulate a hierarchy of approximations based on the size of what we shall call proper "Kohn neighborhoods." We make the idea of these neighborhoods precise and completely work out the results obtained by the use of a drastically truncated version of the smallest such proper Kohn neighborhood.

The results of our analysis are shown to contain all the first-order corrections, in a z^{-1} power expansion, to the mean-field theory, which itself is correct to the dominant term in such a series expansion.

For the case of a sc lattice, we compute the magnetic single-site density of states for several different concentrations. This density of states is shown to conserve the first four frequency moments exactly. All higher-order moments are, of course, given correctly only to the two leading orders in z^{-1}.

Using our results for the magnetic single-site Green's function, we compute the renormalization of the long-wavelength spin-wave energy $E_{\vec{K}}$ as a function of the magnetic concentration. Following Murray,[12] we interpret it to be the same as the renormalization of the Curie temperature $T_C(m)$. Moreover, we plot a graph showing the variation of the real and the imaginary parts of the coherent exchange integral as a function of the frequency for several different concentrations. We also discuss how, from these results, the energy and the width of spin waves of arbitrary wave vector $\vec{K}$ may readily be obtained.

II. FORMULATION

We consider a randomly dilute Heisenberg ferromagnet with nearest-neighbor isotropic exchange interaction acting between magnetic atoms and with vanishingly small external field. The external field is needed only to break the rotational symmetry of the ground state. Once a preferred direction, e.g., the direction of the z axis, has been established, we proceed to the limit of zero field.

As mentioned earlier, in the dense limit the ground state is exactly known.[21] Moreover, in this limit the system thermodynamics is largely that of a gas of noninteracting harmonic-oscillator-like excitations as long as the system temperature T is much smaller than a characteristic Curie temperature T_C, for which the Boltzmann randomizing energy $k_B T_C$ is of the same order of magnitude as the aligning energy caused by exchange interaction between a given atom and its surroundings.

Unlike the dense case, the physics of dilute ferromagnets is not straightforward. Indeed, since the ground state depends upon the precise configuration in which Nm magnetic spins are distributed over a total of N available sites, the computation of the system thermodynamics for the case $m<1$ is far from simple.[1]

The object of the present study is to investigate the properties of the system in the limit of very low temperatures but arbitrary concentration of the magnetic atoms, i.e., $1 \geq m > m_c$. Here m_c is defined as the largest possible concentration of magnetic atoms for which ferromagnetic ground state with long-range magnetic order does not obtain.

Making the usual assumption that the exchange interaction between any pair of magnetic atoms is independent of the presence of other atoms and as such the effective exchange integral between magnetic atoms is not a function of the concentration m, we can write the Hamiltonian for the given configuration as

$$\mathcal{H}^c = -\mu \sum_f{}' S_f^z - \sum_{f_1}{}' \sum_{f_2}{}' I(f_1, f_2)\, \vec{S}_{f_1}\cdot\vec{S}_{f_2} \; . \tag{2.1}$$

As stated earlier, we shall consider the limit for which the Zeeman energy term is vanishing, i.e.,

$$\mu \to +0 \; . \tag{2.2a}$$

Since the exchange integral $I(f_1, f_2)$ acts only between a pair of magnetic atoms at f_1 and f_2, the primed lattice sums in Eq. (2.1) are taken only over magnetic atoms. It is convenient to introduce magnetic occupation operators σ_{f_1}, σ_{f_2}, whereby

$$I(f_1, f_2) = I(f_2, f_1) = J(f_1 f_2)\, \sigma_{f_1}\, \sigma_{f_2} \; . \tag{2.2b}$$

The σ_f's are idempotent operators with eigenvalues $+1$ and 0. The eigenvalue $+1$ occurs only if a magnetic atom is sitting on the position f. Otherwise, the eigenvalue of σ_f is zero. The c-number function $J(f_1 f_2)$ is defined as follows:

$$\begin{aligned} J(f_1 f_2) = J(f_2 f_1) &= J \quad \text{if } f_1 \text{ and } f_2 \text{ are neighboring lattice sites} \\ &= 0 \quad \text{otherwise} \; . \end{aligned} \tag{2.2c}$$

Note that in the definition of $J(f_1 f_2)$ we have made no reference to the species of the atoms sitting on the positions f_1 and f_2. Therefore, in the expression for the configurational Hamiltonian the primed sums over the magnetic sites f_1, f_2 can now be replaced by free sums over the entire lattice, i.e.,

$$\mathcal{H}^c = -\sum_{f_1, f_2} J(f_1 f_2)\, \sigma_{f_1}\, \sigma_{f_2}\, \vec{S}_{f_1}\cdot\vec{S}_{f_2} \; . \tag{2.3}$$

Following Holstein and Primakoff,[21] we define boson spin-deviation creation and destruction operators $a_g^\dagger$, a_g, where

$$[a_g, a_f]_- = [a_g^\dagger, a_f^\dagger]_- = 0 \; , \quad [a_g, a_f^\dagger]_- = \delta_{g,f} \; , \tag{2.4}$$

via the transformation

$$\begin{aligned} S_g^+ &= (2S)^{1/2}(1 - n_g/2S)^{1/2} a_g \; , \\ S_g^- &= (2S)^{1/2} a_g^\dagger (1 - n_g/2S)^{1/2} \; , \\ S_g^z &= S - n_g \; , \end{aligned} \tag{2.5}$$

where

$$n_g = a_g^\dagger a_g \; . \tag{2.6}$$

Because we intend to consider the spin dynamics at very low temperatures, we ignore interactions between the boson excitations by putting

$$(1 - n_g/2S)^{1/2} \sim 1 \; . \tag{2.7}$$

In this so-called noninteracting spin-wave approximation, the magnetic Hamiltonian (2.1) reduces to the following, involving a set of harmonic-oscillator-like excitations distributed over a lattice containing N sites, i.e.,

$$\mathcal{H}^c = 2S \sum_{f_1, f_2} J(f_1 f_2)\, \sigma_{f_1} \sigma_{f_2}\, (n_{f_1} - a_{f_1}^\dagger a_{f_2}) + \text{const.} \tag{2.8}$$

Note that even though we have not placed any site restrictions on the boson operators, because of the presence of the magnetic occupation operators σ_{f_1}, σ_{f_2}, this causes no confusion and only those terms can contribute to $\mathcal{H}^c$ which actually refer to magnetic atoms. While the physical content of the problem is not violated in any way, this freedom of interpretation of the role of the boson operators greatly facilitates the algebraic manipulations to be described in the following sections.

III. GREEN'S FUNCTION

It is convenient to introduce a temperature- and configuration-dependent double-time retarded Green's function with explicit reference to two given sites g and g', i.e.,

$$G_{g,g'}^c(t, t') = -2\pi i\, \Theta(t - t')\, \langle [a_g(t) \, , \; a_{g'}^\dagger(t')]_- \rangle_c \; . \tag{3.1}$$

Here $\Theta(t - t')$ is the usual Heaviside unit step function

$$\Theta(x) = +1 \quad \text{for } x > 0$$

$$= 0 \quad \text{for } x < 0 \; ; \tag{3.2}$$

the time dependence of the operators a_g and $a_g^\dagger$ is in the Heisenberg representation with Dirac units $\hbar = 1$, i.e.,

$$\Omega(t) = e^{it\mathcal{H}^c}\,\Omega(0)\,e^{-it\mathcal{H}^c} , \tag{3.3}$$

and the pointed brackets denote a partial configuration-dependent canonical average, i.e.,

$$\langle \cdots \rangle_c = \frac{\mathrm{Tr}(e^{-\beta\mathcal{H}^c} \ldots)}{\mathrm{Tr}(e^{-\beta\mathcal{H}^c})} \; . \tag{3.4}$$

Note that the trace in Eq. (3.4) is taken over all those states which are consistent with the given configuration in which Nm magnetic and $N(1-m)$ nonmagnetic atoms have been distributed over the N lattice sites. Because no summation over states referring to other configurations occurs, the average $\langle \cdots \rangle_c$ is not the fully thermodynamic average.[1,22]

Note that the system is not subject to time varying external fields, i.e., $\mathcal{H}^c(t)$ commutes with $\mathcal{H}^c(t')$. Consequently, the Green's function (3.1) displays translational invariance with respect to the time difference $(t - t')$, i.e.,

$$G^c_{g,g'}(t, t') = G^c_{g,g'}(t - t') \; . \tag{3.5a}$$

This property enables us to make use of the Fourier transformation:

$$G^c_{g,g'}(t, t') = \int G^c_{g,g'}(E)\, e^{-iE(t-t')}\, dE \; . \tag{3.5b}$$

In contrast with its dependence on time, the dependence of $G^c_{g,g'}(t, t')$ on the locations g and g' is not subject to translational invariance. Rather, for any given configuration, the dependence of the Green's function on the locations g and g' is specific. Consequently, an appropriate Fourier transformation is

$$G^c_{g,g'}(E) = \left(\frac{1}{N}\right)^2 \sum_{\vec{K}} \sum_{\vec{K}'} G^c_{\vec{K},\vec{K}'}(E)\, e^{i(\vec{K}\cdot\vec{g}+\vec{K}'\cdot\vec{g}')} . \tag{3.6}$$

Because of the regularity of the crystal lattice (note this refers only to the spatial locations of the N sites in the lattice), the sums over $\vec{K}$ and $\vec{K}'$ are restricted to the first Brillouin zone which contains a total of N allowed values for each of the inverse lattice vectors $\vec{K}$ and $\vec{K}'$.

Let us now study the equation of motion of this Green's function. Because

$$\frac{i\,da_g(t)}{dt} = [a_g, \mathcal{H}^c]_-$$

$$= 2S\Big[\sum_f \sigma_g \sigma_f J(gf)\Big] a_g - 2S \sum_f \sigma_g \sigma_f J(gf)\, a_f \tag{3.7}$$

and

$$\frac{d\Theta(t-t')}{dt} = \delta(t - t') = \frac{1}{2\pi}\int_{-\infty}^{+\infty} dE\, e^{-iE(t-t')} , \tag{3.8}$$

we readily find

$$E\, G^c_{g,g'}(E) = \delta_{g,g'} + 2S\Big[\sum_f \sigma_g \sigma_f J(gf)\Big] G^c_{g,g'}(E) - 2S\Big[\sum_f \sigma_g \sigma_f J(gf)\Big] G^c_{f,g'}(E) \; . \tag{3.9}$$

Note that the terms on the right-hand side of Eq. (3.9) are explicitly dependent upon the magnetic configuration. For example, if we should be looking at a Green's function for which, say, the site g is known not to be occupied by a magnetic atom, then the last two terms on the right-hand side would be equal to zero and the solution of the Green's function would simply be that for a gas of noninteracting boson excitations of zero energy. Only when the site g is magnetic, the Green's function would have a chance of yielding a nontrivial result.

The Green's-function equation given in Eq. (3.9) describes a set of N^2 simultaneous linear equations. Even though the only nontrivial Green's functions are those for which both the sites g and g' have magnetic atoms, their exact solution for any macroscopically relevant random situation is completely impractical via the well-known elementary algebraic procedures used for solving simultaneous linear equations. Therefore, we have to devise some approximation procedure based upon suitable physical insight.

IV. MATHEMATICAL PRELIMINARIES

Adding a set of terms $A(E)$,

$$A(E) = -2S\sum_f \tilde{J}_{(E)}(gf)\, G^c_{g,g'}(E) + 2S\sum_f \tilde{J}_{(E)}(gf)\, G^c_{f,g'}(E) , \tag{4.1}$$

where $\tilde{J}_{(E)}(gf)$ is for the present to be considered an arbitrary function of E with a simple dependence on the physical separation between sites g and f, i.e.,

$$\tilde{J}_{(E)}(gf) = \tilde{J}_{(E)}(|g - f|) , \tag{4.2}$$

to both sides of Eq. (3.9) we get

$$E\, G^c_{g,g'}(E) - 2S\Big[\sum_f \tilde{J}_{(E)}(gf)\Big] G^c_{g,g'}(E) + 2S\sum_f \tilde{J}_{(E)}(gf)\, G^c_{f,g'}(E) - \delta_{g,g'}$$

$$= 2S\sum_f \big[J(gf)\,\sigma_f\sigma_g - \tilde{J}_{(E)}(gf)\big]\, G^c_{g,g'}(E) - 2S\sum_f \big[J(gf)\,\sigma_f\sigma_g - \tilde{J}_{(E)}(gf)\big]\, G^c_{f,g'}(E) \; . \tag{4.3}$$

This equation is identical to and contains exactly the same information as Eq. (3.9). Our objective in writing it in the given form, however, is to evolve a procedure which can be used for the calculation of magnetic single-site Green's function in thermal equilibrium. As we shall show later, this can be achieved by the device of making the right-hand side of Eq. (4.3) appropriately small for the case of thermal equilibrium. It is hoped that this would in turn lead to some self-consistent physical criteria for the choice of the function $\tilde{J}_{(E)}(f_1 f_2)$.

To this end, let us begin by using the following compact notation:

$$V^c_{g,p}(E) = \delta_{g,p} 2S \sum_f [J(gf)\sigma_g\sigma_f - \tilde{J}_{(E)}(gf)] - 2S[J(gp)\sigma_g\sigma_p - \tilde{J}_{(E)}(gp)] \ . \tag{4.4}$$

With this notation, Eq. (4.3) becomes

$$E\, G^c_{g,g'}(E) - 2S[\sum_f \tilde{J}_{(E)}(gf)]\, G^c_{g,g'}(E) + 2S\sum_f \tilde{J}_{(E)}(gf)\, G^c_{f,g'}(E) - \delta_{g,g'} = \sum_p V^c_{gp}(E)\, G^c_{p,g'}(E) \ . \tag{4.5}$$

Like $G^c_{g,g'}(E)$ the dependence of $V^c_{g,p}(E)$ is specific to sites g and p. On the other hand, $\tilde{J}_{(E)}(gf)$ and $\delta_{g,f}$ are functions only of the separation $|g-f|$. Therefore an appropriate Fourier transformation is as follows:

$$V^c_{g,p}(E) = \left(\frac{1}{N}\right)^2 \sum_{\vec{K}_1}\sum_{\vec{K}_2} V^c_{\vec{K}_1,\vec{K}_2}(E)\, e^{i(\vec{K}_1\cdot g + \vec{K}_2\cdot p)} \ , \tag{4.6}$$

$$\tilde{J}_{(E)}(gp) = \frac{1}{N}\sum_{\vec{K}} \tilde{j}_{(E)}(\vec{K})\, e^{i\vec{K}\cdot(\vec{g}-\vec{p})} \ , \tag{4.7}$$

$$\delta_{g,p} = \frac{1}{N}\sum_{\vec{K}} e^{i\vec{K}\cdot(\vec{g}-\vec{p})} \ . \tag{4.8}$$

[Compare Eqs. (3.6) and (4.6).]

Introducing the transformations (3.6), (4.6)–(4.8), into Eq. (4.5) we get

$$G^c_{\vec{K}_1,\vec{K}_2}(E)[E - 2S\tilde{j}_{(E)}(0) + 2S\tilde{j}_{(E)}(\vec{K}_1)] - N\delta_{\vec{K}_1,-\vec{K}_2} = \frac{1}{N}\sum_{\vec{K}} V^c_{\vec{K}_1,-\vec{K}}(E)\, G^c_{\vec{K},\vec{K}_2}(E) \ .$$

Upon multiplying both sides of this equation by $N^{-2}[E - 2S\tilde{j}_{(E)}(0) + 2S\tilde{j}_{(E)}(\vec{K}_1)]^{-1} e^{i(\vec{K}_1 g + \vec{K}_2 g')}$ and summing over all the inverse lattice vectors $\vec{K}_1$, $\vec{K}_2$ contained within the first Brillouin zone, we get

$$G^c_{g,g'}(E) = \Gamma_{g,g'}(E) + \sum_{l,p} \Gamma_{g,l}(E)\, V^c_{l,p}(E)\, G_{p,g'}(E) \ , \tag{4.9}$$

where

$$\Gamma_{g,g'}(E) = \frac{1}{N}\sum_{\vec{K}} \frac{e^{i\vec{K}(g-g')}}{E - 2S\tilde{j}_{(E)}(0) + 2S\tilde{j}_{(E)}(\vec{K})} \ . \tag{4.10}$$

The significance of this equation lies in the fact that while $G^c_{g,g'}(E)$ is a Green's function which depends explicitly on the given configuration in which Nm magnetic atoms and $N(1-m)$ nonmagnetic atoms are distributed over the N lattice sites, the Green's function $\Gamma_{g,g'}(E)$ has no such dependence on the configuration. Rather, it depends on the vector separation $(g-g')$ and is a function only of the quantity $\tilde{J}_{(E)}(f_1 f_2)$ which is still arbitrary. The entire configurational dependence of $G^c_{g,g'}(E)$ has thus been transferred to $V^c_{l,p}(E)$ which appears on the right-hand side of Eq. (4.9).

[Note that although the development of this section is somewhat parallel to that used in the coherent potential approximation theories[23,24] for studying the dynamics of electronic disorder, the one-to-one correspondence of the on-site potentials with the off-diagonal hopping interactions that occurs in Eq. (4.4) is unique to the isotropic Heisenberg spin systems.]

V. *T* MATRIX

We are now in a position to describe the scattering T matrix. Let us first introduce a set of three space- and energy-dependent matrices, i.e.,

$$G^c(E) \equiv [G^c_{i,j}(E)] \ ,$$

$$V^c(E) \equiv [V^c_{i,j}(E)] \ ,$$

$$\Gamma(E) \equiv [\Gamma_{i,j}(E)] \ . \tag{5.1}$$

The matrix elements of these E-dependent matrices are taken with respect to the two lattice positions, for example, i and j. Therefore, these are $N\times N$ square matrices.

With this notation, Eq. (4.9) becomes an arbitrary matrix element of the following matrix equation:

$$G^c(E) = \Gamma(E) + \Gamma(E)\, V^c(E)\, G^c(E) \ . \tag{5.2}$$

Now, if we define a new $N\times N$ square matrix in the same, lattice-site index, notation through the relationship

$$V^c(E)\, G^c(E) \equiv T^c(E)\, \Gamma(E) \ , \tag{5.3}$$

then Eq. (5.2) becomes

$$G^c(E) = \Gamma(E) + \Gamma(E)\, T^c(E)\, \Gamma(E) \ , \tag{5.4}$$

and, moreover, the matrix $T(E)$ satisfies the equation

$$T^c(E) = V^c(E) + V^c(E)\, \Gamma(E)\, T^c(E) \ . \tag{5.5}$$

[To derive Eq. (5.5), multiply both sides of Eq. (4.12) from the left with the matrix $V^c(E)$ and replace $V^c(E)\,G^c(E)$ by $T^c(E)\,\Gamma(E)$ on its right-hand side. Because $\Gamma(E)$ is arbitrary, the resultant equation leads to Eq. (5.5).]

It should be emphasized here that the introduction of the T matrix has by itself not brought us any closer to the exact solution of the Green's function because the full configurational complexity has merely been transferred to the T matrix from the scattering potential matrix $V^c(E)$. On the other hand, inasmuch as the T matrix scatters approximately renormalized excitations propagated by $\Gamma(E)$, it is more susceptible to approximation procedures based upon the use of physical insight into the structure of the many-body dynamics than is the configurational Green's function itself.

Kohn[20] has argued that the dynamical properties of a sufficiently random many-body system are well approximated by the actual situation prevailing in small neighborhoods. Loosely stated, his assertion is that for any given frequency ω, if the size of a typical small neighborhood is characterized by a length R, then the errors involved in equating the dynamical properties of the large thermodynamic system with those of the typical small neighborhood decrease exponentially with the increase in the ratio $R/\lambda(\omega)$. Here $\lambda(\omega)$ is some typical mean free path for frequency ω.

In this paper we intend to make use of this compelling physical argument to derive an approximation procedure for computing the magnetic self-Green's function. To this end, let us assume that a reasonable approximation to the system dynamics, over much of the frequency range, would be obtained if we equated its properties with that of a small Kohn neighborhood whose size is typified by a length R_n (where R_n is equal to the distance between nth nearest neighbors in the lattice). To examine the consequences of such an assumption, let us take matrix elements of the two sides of Eq. (5.5) between a site i and an arbitrary central site 0, i.e.,

$$T^c_{i,0}(E) = V^c_{i,0}(E) + [V^c(E)\,\Gamma(E)\,T^c(E)]_{i,0}\ . \qquad (5.6)$$

Then, according to Kohn's philosophy, if i lies within a distance R_n to the site 0, then it would be considered to be a member of the neighborhood and should as such be taken into account explicitly. However, if i lies farther than R_n, then its influence on the system properties should be considered negligible.

We extend these ideas by proposing a somewhat different, and in our view more meaningful, operational routine. This routine asserts that all those matrix elements of the $T^c(E)$ matrix, as well as those of the renormalized translationally invariant Green's-function matrix $\Gamma(E)$ (for which the mutual separation of the two lattice sites that the matrix elements refer to is larger than R_n) are to be ignored in comparison with those for which the corresponding separation is shorter than R_n. The system dynamics within this truncation procedure, in analogy with Kohn's philosophy, is then expected to approximate the dynamics of the large thermodynamic system to an accuracy which can be made systematically greater by increasing the range R_n.

With this assumption the simultaneous linear equations for $T^c_{i,0}(E)$, given in Eq. (5.6), truncate and form a closed set. Their solution is, therefore, elementary, albeit very tedious, depending upon the crystal structure and the spatial range R_n of the matrix elements of $T^c(E)$ and $\Gamma(E)$.

After the various matrix elements $T^c_{i,0}(E)$ have thus been determined from the solution of Eq. (5.6), our job would be to compute the Green's function. Clearly, the Green's function of interest is not the configuration-dependent Green's function $G^c_{g,g'}(E)$, but rather the thermodynamic average of these Green's functions over all possible configurations, i.e., $\overline{G}_{g,g'}(E)$, where

$$\overline{G}_{g,g'}(E) = \frac{\mathrm{Tr}[G^c_{g,g'}(E)]}{\mathrm{Tr}[1]}\ . \qquad (5.7)$$

The traces in Eq. (5.7) are taken over all possible configurations.[1,22] [Note that the thermodynamic Green's function, unlike $G^c_{g,g'}(E)$, will show translational invariance, i.e., it will be a function only of the vector separation $(g-g')$, because in thermodynamic equilibrium all sites are equivalent.]

The computation of $\overline{G}_{g,g'}(E)$ can now proceed straightforwardly. Taking matrix elements of Eq. (5.4) with respect to positions g and g', and taking the trace over all configurations we find

$$\overline{G}_{g,g'}(E) = \Gamma_{g,g'}(E) + \sum_{l,l'} \Gamma_{g,l}(E)\,\overline{T}_{l,l'}(E)\,\Gamma_{l',g'}(E)\ . \qquad (5.8)$$

If we could now arrange to make the term containing $\overline{T}_{l,l'}(E)$, occurring on the right-hand side of Eq. (5.8), to be vanishingly small, i.e.,

$$\overline{T}_{l,l'}(E) = 0 \quad \text{for} \quad |l-l'| \leqslant R_n\ , \qquad (5.9\text{a})$$

then subject to the errors inherent in the restricted-range assumption, i.e.,

$$\overline{T}_{l,l'}(E) \sim 0 \quad \text{for} \quad |l-l'| > R_n\ , \qquad (5.9\text{b})$$

we would have

$$\overline{G}_{g,g'}(E) \simeq \Gamma_{g,g'}(E)\ . \qquad (5.10)$$

Because the choice of the function $\tilde{J}_{(E)}(f_1 f_2)$ is still open, the second term on the right-hand side of Eq. (5.8) can therefore be made to vanish by a suitable choice of this function.

It is worthwhile here to point out that the theory presented so far applies with minor modifications to the case of electronic alloys with random off-diagonal hopping or to the case where the external magnetic field μ is also present. However, when we specialize to spin systems for the simple case of $\mu = 0$, we can make use of an exact sum rule. To derive this sum rule, let us sum the matrix elements $V^c_{g,p}(E)$ given in Eq. (4.4) over all the sites g in the lattice, or over all the sites p in the lattice. We get

$$\sum_g V^c_{g,p}(E) = \sum_p V^c_{g,p}(E) = 0 . \tag{5.11}$$

Now if we sum both sides of Eq. (5.6) over all the sites i in the lattice, we get

$$\sum_i T^c_{i,0}(E) = \sum_i V^c_{i,0}(E) + \sum_l \Big[\sum_i V^c_{i,l}(E)\Big] \times \Big[\sum_{l'} \Gamma_{l,l'}(E)\, T^c_{l',0}(E)\Big] . \tag{5.12}$$

Using (5.11) in (5.12), we get the relevant sum rule

$$\sum_i T^c_{i,0}(E) = 0 . \tag{5.13}$$

As a consequence of this sum rule, we have a total of only n unknowns for the case where the range of the matrix elements of $T^c(E)$ and $\Gamma(E)$ is supposed to be equal to the separation of nth nearest neighbors in the lattice. These unknowns are determined through the choice of $\tilde{J}_{(E)}(|g-p|)$:

$$|g-p| = R_\nu, \quad \nu = 1, 2, \ldots, n \tag{5.14}$$

such that the following n equations are satisfied:

$$\overline{T}_{i,0}(E) = 0 \quad \text{for } |i-0| \leq R_n . \tag{5.15}$$

VI. NEAREST-NEIGHBOR CLUSTER

The theory presented in Sec. V was rather general. A clearer understanding of the mechanics of the present approximation procedure is achieved by considering a specific-sized Kohn neighborhood. Let us, to begin with, inquire into what happens if we were to limit the spatial range of the matrix elements of $T^c(E)$ and $\Gamma(E)$ to the separation of nearest neighbors. In this case, the operational routine would first be to put $T^c_{i,0}(E) \simeq 0$ whenever the separation of i and 0 exceeded the nearest-neighbor distance. Then Eqs. (5.6) and (5.13) would be used to give

$$T^c_{0,0}(E) = -T^c_{1,0}(E) - \cdots - T^c_{z,0}(E)$$

$$= V^c_{0,0}(E) + [V^c(E)\,\Gamma(E)]_{00}\, T^c_{0,0}(E) + [V^c(E)\,\Gamma(E)]_{01}\, T^c_{1,0}(E) + \cdots + [V^c(E)\,\Gamma(E)]_{0z}\, T^c_{z,0}(E) , \tag{6.1}$$

$$T^c_{n,0}(E) = V^c_{n,0}(E) + [V^c(E)\,\Gamma(E)]_{nn}\, T^c_{n,0} + [V^c(E)\,\Gamma(E)]_{n0}\, T^c_{0,0}(E) + \sum_{p\neq 0;\, p\neq n} [V^c(E)\,\Gamma(E)]_{np}\, T^c_{p,0}(E) . \tag{6.2}$$

The matrix elements $T^c_{n,0}(E)$, $T^c_{p,0}(E)$ denote matrix elements between the site 0 and the site n or p. Here n and p are any of the z nearest neighbors of the site 0. Note that only after the configurational average of the matrix elements like $T^c_{n,0}(E)$ has been taken can we expect these matrix elements to be equal. Without such averaging, they are in general all different, being explicitly dependent upon the composition of an environment of the site 0 that we shall now describe.

For a one-dimensional lattice, Eqs. (6.1) and (6.2) reduce to two equations in the unknowns $T^c_{1,0}(E)$ and $T^c_{2,0}(E)$. (Here 1 and 2 denote the nearest neighbors of the site 0.) Solving these two simultaneous linear equations gives us $T^c_{1,0}(E)$ and $T^c_{2,0}(E)$, in terms of the properties of a set of five lattice sites labeled 0, 1, 2, 3, 4 [see Fig. 1(a)]. By fulfilling the requirement that the configurational average $\overline{T}_{1,0}(E)$ is vanishing, i.e.,

$$\overline{T}_{1,0} = \mathrm{Tr}[T^c_{1,0}(E)]/\mathrm{Tr}[1] , \tag{6.3}$$

we automatically ensure that

$$\overline{T}_{2,0} = 0 ; \quad \overline{T}_{0,0}(E) = \overline{T}_{1,0}(E) + \overline{T}_{2,0}(E) = 0 . \tag{6.4}$$

[Note that the trace in Eq. (6.3) need only be taken over the occupation states of the sites 0–4.] Thus, there is only one unknown to be determined here which we shall simply call $\tilde{J}_{(E)}$, and it is determined by Eq. (6.3). Note that since Eq. (6.3) takes a configurational average over the occupation states of five sites, it contains a total of $(2)^5$ terms.

In a two-dimensional square lattice [see Fig. 1(b)], Eqs. (6.1) and (6.2) involve four unknowns $T^c_{1,0}$, $T^c_{2,0}$, $T^c_{3,0}$, and $T^c_{4,0}$. Obtaining the solution for any one of these, say $T^c_{1,0}$, and using the requirement that its configurational average be vanishing, ensures that the configurational averages of all the five matrix elements $T^c_{0,0}$, $T^c_{1,0}, \ldots, T^c_{4,0}$ will be zero. Note that this time the configurational averaging is more tedious because it explicitly involves 13 sites and as such has $(2)^{13}$ terms in it.

In a simple three-dimensional lattice (i.e., simple cubic), Eqs. (6.1) and (6.2) lead to six coupled equations for $T^c_{1,0}$, $T^c_{2,0}$, etc. The solution for $T^c_{1,0}$, and the subsequent imposition of the requirement that its configurational average be vanishing would ensure that the configurational average of all seven matrix elements $T^c_{0,0}$, $T^c_{1,0}, \ldots, T^c_{6,0}$ is vanishing. Unfortunately, however, any of these matrix elements, e.g., $T^c_{1,0}$, contains explicit refer-

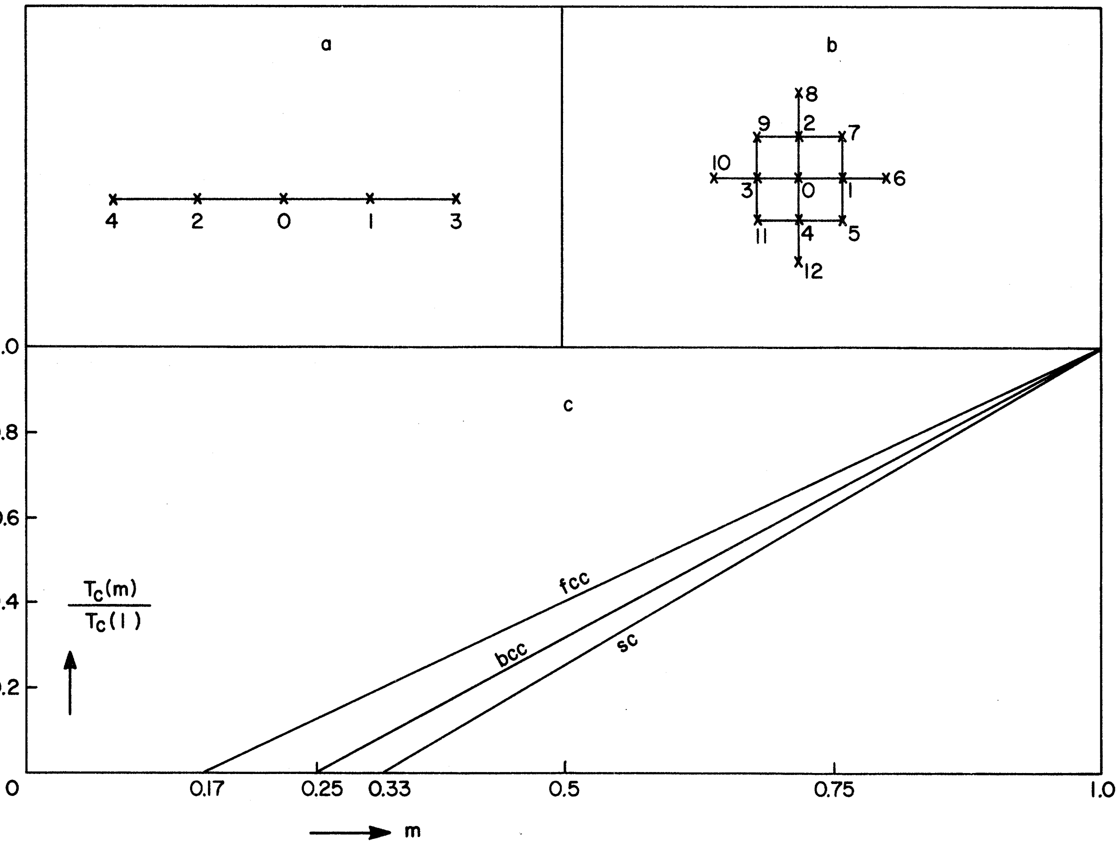

FIG. 1. (a) and (b) Proper nearest-neighbor Kohn neighborhoods, centered around a site 0, in linear and quadratic square lattices. (c) Renormalization of the $K\rightarrow 0$ spin-wave energy as a function of the magnetic concentration for cubic lattices.

ence to 25 sites, and therefore, the equation describing the configurational average of $T^c_{1,0}$ will contain a maximum of $(2)^{25}$ terms.

Assuming that the foregoing procedure has been followed and due to an appropriate choice of $\tilde{J}_{(E)}$, the configurational averages of the nearest-neighbor matrix elements, e.g., $\overline{T}_{1,0}(E)$, have been self-consistently equated to zero, then, subject to the accuracy of the given Kohn neighborhood approximation [i.e., whereby matrix elements $T^c_{i,j}(E)$ and $\Gamma_{i,j}(E)$ are assumed to be vanishingly small whenever the separation of i and j is greater than the nearest-neighbor distance] we get

$$\begin{aligned}\overline{G}_{g,g'}(E) &= \Gamma_{g,g'}(E)+\sum_{l,l'}\Gamma_{g,l}(E)\,\overline{T}_{l,l'}(E)\,\Gamma_{l',g}(E)\\ &\simeq \Gamma_{g,g'}(E)\ \text{for}\ |g-g'|\leqslant R_1\\ &\simeq 0\ \text{for}\ |g-g'|>R_1\,. \qquad (6.5)\end{aligned}$$

Before we conclude this section, it is helpful to get a feel for the accuracy that the nearest-neighbor Kohn-cluster solution outlined here would have. In self-consistent theories, the preservation of the first several frequency moments of the dynamical spectral function is traditionally considered to be a useful guide to their inherent accuracy.[24] As will be described in Sec. VIII, the exact computation of the first several frequency moments is straightforward and upon examining their structure it is possible to predict the number of frequency moments the nearest-neighbor cluster will preserve. These considerations lead us to believe that in lattices without nearest-neighbor triangles the self-consistent solution to the nearest-neighbor cluster theory outlined here will exactly preserve the first six moments, $\overline{M}_0$ to $\overline{M}_5$, of the average single-site density of states $\overline{\rho}(\omega)$, where

$$\overline{M}_\nu=\int_{-\infty}^{+\infty}\overline{\rho}(\omega)\,\omega^\nu\,d\omega\,, \qquad (6.6)$$

$$\begin{aligned}\overline{\rho}(\omega) &= \frac{1}{2\pi}\int_{-\infty}^{+\infty}\langle[a_g(t),\ a_g^\dagger(t')]_-\rangle_{\text{thermal}}\,e^{i\omega(t-t')}\,d(t-t')\\ &= -\frac{1}{\pi}\,\text{Im}\overline{G}_{g,g'}(\omega+i\epsilon)\,, \qquad \epsilon\rightarrow+0\,. \qquad (6.7)\end{aligned}$$

Note that in Eq. (6.7), g is any arbitrary site in the lattice. Moreover, it should be noted that the average single-site density of states $\bar{\rho}(\omega)$ is related to the magnetic site density of states $\rho_m(\omega)$ and the nonmagnetic site density of states $\rho_{n.m}(\omega)$ through the following relation which obtains for complete substitutional disorder:

$$\bar{\rho}(\omega) = m\,\rho_m(\omega) + (1-m)\,\rho_{n.m}(\omega)\ . \tag{6.8}$$

(The significance of the absence of the bar on the magnetic and the nonmagnetic densities of states will be made clear in Sec. VII.)

VII. A PAIR APPROXIMATION

In three dimensions, the self-consistent determination of $\tilde{J}_{(E)}$, in the nearest-neighbor cluster approximation described in Sec. VI, would require the use of Eq. (6.4) which can have a maximum of $(2)^{25}$ terms. This is clearly too tedious a calculation to be undertaken analytically. The development of a computer program for generating, classifying, and summing these $(2)^{25}$ terms is currently underway and will be described at some future date.[25] For the present, we content ourselves with a truncated nearest-neighbor approximation involving only two neighboring sites. Because such an approximation refers explicitly only to two sites, the relevant configurational average involves a maximum of only four terms.

Consider two arbitrary neighboring sites labeled 0 and 1, and ignore the presence of all other sites in the nearest-neighbor Kohn neighborhood described in Sec. VI. Let us write the truncated version of Eqs. (6.1) and (6.2), referring to such a two-site neighborhood. We have

$$\begin{aligned} T^c_{0,0}(E)\,[1 - V^c_{0,0}(E)\,\Gamma_{0,0}(E) - V^c_{0,1}(E)\,\Gamma_{1,0}(E)] \\ = V^c_{0,0}(E) + [V^c_{0,0}(E)\,\Gamma_{0,1}(E) \\ + V^c_{0,1}(E)\,\Gamma_{1,1}(E)]\,T^c_{1,0}(E)\ , \end{aligned} \tag{7.1}$$

$$\begin{aligned} T^c_{1,0}(E)\,[1 - V^c_{1,1}(E)\,\Gamma_{1,1}(E) - V^c_{1,0}(E)\,\Gamma_{0,1}(E)] \\ = V^c_{1,0}(E) + [V^c_{1,0}(E)\,\Gamma_{0,0}(E) \\ + V^c_{1,1}(E)\,\Gamma_{1,0}(E)]\,T^c_{0,0}(E)\ . \end{aligned} \tag{7.2}$$

Because the Green's function $\Gamma_{i,j}(E)$ is symmetric with respect to spatial inversions and translations (note it only depends on $\tilde{J}_{(E)}$), we have

$$\Gamma_{0,0}(E) = \Gamma_{1,1}(E) \equiv \Gamma^{(0)}(E)\ , \tag{7.3}$$

$$\Gamma_{0,1}(E) = \Gamma_{1,0}(E) \equiv \Gamma^{(1)}(E)\ . \tag{7.4}$$

Moreover, from Eq. (4.4) it follows that for such a truncated two-site neighborhood

$$V^c_{0,1}(E) = V^c_{1,0}(E) = -2S(\sigma_0\sigma_1 J - \tilde{J}_{(E)})\ , \tag{7.5}$$

$$V^c_{0,0}(E) = V^c_{1,1}(E) = -V^c_{0,1}(E)\ . \tag{7.6}$$

Using Eqs. (7.3)–(7.6), Eqs. (7.1) and (7.2) readily yield the result

$$\begin{aligned} T^c_{0,0}(E) &= V^c_{0,0}(E)\,\{1 - 2\,V^c_{0,0}(E)\,[\Gamma^{(0)}(E) - \Gamma^{(1)}(E)]\}^{-1} \\ &= T^c_{1,1}(E) = -T^c_{1,0}(E) = -T^c_{0,1}(E)\ . \end{aligned} \tag{7.7}$$

Having thus determined all the matrix elements for the two-site neighborhood, our next task is to invent a suitable averaging procedure such that it leads to a reasonable choice for the function $\tilde{J}_{(E)}$. However, before we attempt this, we must keep in mind the fact that the thermodynamic averaging procedure described in Sec. VI is designed to work only when a consistent choice for the Kohn neighborhood is made. Because the two-site truncation contained in Eqs. (7.1)–(7.7) is not consistent in the sense made explicit in Sec. VI, the following procedure is more accurate to use here.

Instead of working with the thermodynamic Green's function $\bar{G}_{g,g'}(E)$, let us make use of the magnetic Green's function $G_{0,0}(E)$. This Green's function is defined by using the conditional average:

$$G_{0,0}(E) = \mathrm{Tr}[\sigma_0\, G^c_{0,0}(E)]/\mathrm{Tr}(\sigma_0)\ . \tag{7.8}$$

[Note the absence of the superscript c and the bar from $G_{0,0}(E)$.] Although the trace in Eq. (7.8) is taken over all configurations, the presence of the magnetic occupation operator ensures that only those terms contribute which are consistent with the presence of a magnetic atom on the site 0. Because

$$G^c_{0,0}(E) = \Gamma_{0,0}(E) + [\Gamma(E)\,T^c(E)\,\Gamma(E)]_{0,0}\ , \tag{7.9}$$

for the truncated neighborhood consisting only of sites 0 and 1 we have

$$\begin{aligned} G_{0,0}(E) = \Gamma^{(0)}(E) + [\Gamma^{(0)}(E)]^2\,T_{0,0}(E) + \Gamma^{(0)}(E) \\ \times \Gamma^{(1)}(E)\,[T_{1,0}(E) + T_{0,1}(E)] \\ + [\Gamma^{(1)}(E)]^2\,T_{1,1}(E)\ , \end{aligned} \tag{7.10}$$

where the absence of any superscript on $G_{0,0}(E)$ and the matrix elements of $T(E)$ indicates a conditional averaging over the truncated neighborhood, i.e., for sites $i=0$, 1 and $j=0$, 1,

$$\Omega_{i,j}(E) = \mathrm{Tr}[\sigma_0\,\Omega^c_{i,j}(E)]/\mathrm{Tr}(\sigma_0)\ . \tag{7.11}$$

All other notation in Eqs. (7.9)–(7.11) is the same as that used in Eqs. (7.3) and (7.4).

In Eq. (7.10), if we now require that for all E,

$$T_{1,0}(E) = 0\ , \tag{7.12}$$

which in turn ensures that

$$T_{0,1}(E) = 0 = -T_{0,0}(E) = -T_{1,1}(E)\ , \tag{7.13}$$

then we get

$$G_{0,0}(E)=\Gamma^{(0)}(E)=\left(\frac{1}{N}\right)\sum_{\vec{K}}\frac{1}{E-2Sz\tilde{J}_{(E)}(1-\gamma_{\vec{K}})}\ , \tag{7.14a}$$

where $\gamma_{\vec{K}}$ is equal to $\cos K_x$ for the linear lattice, $\frac{1}{2}(\cos K_x+\cos K_y)$ for the quadratic lattice, and for three-dimensional cubic lattices

$$\begin{aligned}\gamma_{\vec{K}}&=\tfrac{1}{3}\ (\cos K_x+\cos K_y+\cos K_z) && \text{sc}\\ &=\cos(\tfrac{1}{2}K_x)\cos(\tfrac{1}{2}K_y)\cos(\tfrac{1}{2}K_z) && \text{bcc}\\ &=\tfrac{1}{3}[\cos\tfrac{1}{2}(K_x-K_y)+\cos\tfrac{1}{2}(K_y-K_z)\\ &\qquad+\cos\tfrac{1}{2}(K_z-K_x)] && \text{fcc}\ .\end{aligned} \tag{7.14b}$$

Note that the requirement (7.12) makes use of a conditional average of the Green's function $G^c_{0,0}(E)$, and as such is much more stringent than the corresponding requirement discussed in Sec. VI [compare Eqs. (6.3) and (6.4), which are relevant to one dimension, with Eq. (7.12) which refers to general lattice dimensionality]. The reason for this is that the two-site neighborhood is a considerably truncated version of the full Kohn neighborhood. While the stronger condition requiring the vanishing of the conditionally averaged matrix elements of $T(E)$ cannot be used in the case of the full nearest-neighbor Kohn neighborhood for the reason that it does not cause the vanishing of the terms $[\Gamma(E)\,T(E)\,\Gamma(E)]_{0,0}$, it is applicable to the truncated two-site neighborhood being discussed in this section. [See Eqs. (7.9) and (7.10).] Note that for this neighborhood, the weaker requirement of the form given in Eqs. (6.3) and (6.4) can also be used, but we anticipate that unlike in the case of the full nearest-neighbor Kohn neighborhood, the use of the weaker requirement will lead to unacceptably large errors in the results for the density of states.

To write Eq. (7.12) more explicitly, we remind ourselves that because the site 0 is occupied by a magnetic atom, the conditional averaging is only taken over the two occupational states of the site 1. Therefore, inserting Eq. (7.7) into (7.12) gives

$$\frac{2S(J-\tilde{J}_{(E)})m}{1-4S(J-\tilde{J}_{(E)})[\Gamma^{(0)}(E)-\Gamma^{(1)}(E)]}+\frac{2S(-\tilde{J}_{(E)})(1-m)}{1-4S(-\tilde{J}_{(E)})[\Gamma^{(0)}(E)-\Gamma^{(1)}(E)]}=0\ . \tag{7.15}$$

The first term on the left-hand side of Eq. (7.15) is contributed by the magnetic occupation of site 1, whereas the second term is contributed by the nonmagnetic occupation of site 1. Because

$$\begin{aligned}\Gamma^{(1)}(E)&=\frac{1}{N}\sum_{\vec{K}}\frac{\gamma_{\vec{K}}}{E-2Sz\tilde{J}_{(E)}(1-\gamma_{\vec{K}})}\\ &=\Gamma^{(0)}(E)-\frac{E\,\Gamma^{(0)}(E)-1}{2Sz\tilde{J}_{(E)}}\ ,\end{aligned} \tag{7.16}$$

Eq. (7.15) reduces to an implicit transcendental equation relating $\tilde{J}_{(E)}$ to $\Gamma^{(0)}(E)$. For three dimensions, its solution can only be obtained numerically as long as $E\neq 0$. For $E=+i0$, however, its solution is straightforward as shown below.

From Eq. (7.16) we have

$$\lim\,[\Gamma^{(0)}(E)-\Gamma^{(1)}(E)]=-(2Sz\tilde{J}_{(i0)})^{-1}\ ,\quad E\to+i0 \tag{7.17}$$

therefore, Eq. (7.15) gives

$$\tilde{J}_{(i0)}=J(mz-2)/(z-2)\ . \tag{7.18}$$

This is an important result in that it expresses the renormalization of the exchange integral for excitations of infinitely long wavelength. This interpretation is based on the fact that the structure of the self-Green's function for $E\to+i0$ must describe the behavior of excitations of vanishingly small energy. In other words, for infintesimal wave vectors the spin-wave energy $E_{\vec{K}}$ is

$$\lim E_{\vec{K}}=\tfrac{2}{6}\,Sz\,\tilde{J}_{(i0)}K^2\ ,\quad K\to 0\ . \tag{7.19}$$

[Here the nearest-neighbor distance is chosen to be the unit of length.] Because the existence of long-wavelength spin waves indicates the presence of magnetic long-range order,[12] from Eqs. (7.18) and (7.19) it follows that when the concentration of magnetic atoms reaches a minimum critical value m_c,

$$m_c=2/z\ , \tag{7.20}$$

then the magnetic long-range order disappears. In cubic lattices the condition (7.19) gives

$$m_c=0.33\ \text{sc}\ ;\quad 0.25\ \text{bcc}\ ;\quad 0.17\ \text{fcc}\ . \tag{7.21}$$

These estimates for the critical concentrations are in good agreement with those given by reliable static theories.[1-11,13,14] Moreover, for the simple-cubic case, where dynamical estimates for m_c, i.e., $m_c=0.34$ and 0.36, are also available,[18] our result 0.33 is in good agreement with them. Indeed, our result (7.18) giving the renormalization of $\tilde{J}_{(i0)}$ with the magnetic concentration is, in general, in fairly good agreement with the corresponding results given by these theories for much of the range $1\geqslant m\geqslant m_c$. Minor discrepancies between our results [plotted in Fig. 1(c)] and those given by the static theories, however, arise in the neighborhood of the critical concentration m_c, whereas the dynamical-theory results for the simple-cubic case remain essentially identical, i.e., to within 3% or so, with ours throughout the ferromagnetic region. [Note that the plot in Fig. 1(c) makes the usual assumption[12] that the ratio of the Curie temperatures of the dilute and the dense system, i.e., $T_C(m)/T_C(1)$, is proportional to the ratio $\tilde{J}_{(i0)}/J$.]

VIII. MAGNETIC SITE DENSITY OF STATE AND SPIN-WAVE SPECTRUM

For arbitrary energies E the transcendental relationship (7.15), which determines $\tilde{J}_{(E)}$, cannot be solved by analytical means for three-dimensional lattices. We have, therefore, used a numerical procedure to compute $\tilde{J}_{(E)}$ for a simple-cubic lattice. This procedure makes use of the well-known[26] representation of $\Gamma^{(0)}(E)$ in terms of the Bessel function of zero order, $J_0(t)$, i.e.,

$$\Gamma^{(0)}(E)=\left(\frac{-3i}{12S\tilde{J}_{(E)}}\right)\int_0^\infty dt\,[J_0(t)]^3 \times\exp[-it(12S\tilde{J}_{(E)}-E)/4S\tilde{J}_{(E)}]\,, \qquad (8.1)$$

and solves for $\tilde{J}_{(E)}$ through the use of Newton-Raphson iteration procedure such that the absolute value of the left-hand side of Eq. (7.15) becomes self-consistently small, i.e., $< 10^{-5}$. Now using this result for $\tilde{J}_{(E)}$ into the right-hand side of Eq. (7.14a) for $E=\omega+i\epsilon$ and proceeding to the limit $\epsilon < 10^{-5}$ we get a numerical estimate for the magnetic single-site density of states $\rho_m(\omega)$,

$$\rho_m(\omega) = -\frac{1}{\pi}\,\mathrm{Im}G_{0,0}(\omega+i\epsilon)\ \ \lim \epsilon\to+0$$

$$\simeq -\frac{1}{\pi}\,\mathrm{Im}\Gamma^{(0)}(E)\ \ \text{for } E=\omega+i\,10^{-5}\,. \qquad (8.2)$$

Before we describe this density of states, it is interesting to introduce the concept of magnetic single-site spin-wave spectrum with energy $E_{\vec{K}}(\omega)$ and width $\psi_{\vec{K}}(\omega)$ through the relations

$$E_{\vec{K}}(\omega) = \mathrm{Re}[2S\tilde{J}_{(\omega+i\epsilon)}(1-\gamma_{\vec{K}})]\,, \quad \epsilon\to+0 \qquad (8.3)$$

$$\psi_{\vec{K}}(\omega) = -\mathrm{Im}[2S\tilde{J}_{(\omega+i\epsilon)}(1-\gamma_{\vec{K}})], \quad \epsilon\to+i0. \qquad (8.4)$$

Because the resultant $\vec{K}$-dependent spectral intensity $\rho_m(\vec{K},\omega)$,

$$\rho_m(K,\omega) = -\frac{1}{\pi}\,\mathrm{Im}G_{\vec{K}}(\omega+i\epsilon)\ \ \epsilon\to+0$$

$$=\frac{1}{\pi}\,\frac{\psi_{\vec{K}}(\omega)}{[\omega-E_{\vec{K}}(\omega)]^2+[\psi_{\vec{K}}(\omega)]^2}\,, \qquad (8.5)$$

where

$$G_{\vec{K}}(E)=[E-2Sz\tilde{J}_{(E)}(1-\gamma_{\vec{K}})]^{-1}\,, \qquad (8.6)$$

$$\rho_m(\omega)=\frac{1}{N}\sum_{\vec{K}}\rho_m(\vec{K},\omega)\,, \qquad (8.7)$$

is a peaked function of frequency ω or the wave vector $\vec{K}$ near the points

$$\omega\sim\tilde{E}_{\vec{K}}(\omega)\,, \qquad (8.8)$$

it is useful to call $\tilde{E}_{\vec{K}}(\tilde{E}_{\vec{K}})$ the spin-wave energy and $\psi_{\vec{K}}(\tilde{E}_{\vec{K}})$ its width. We emphasize that these magnetic single-site spin waves are not identical to the actual $\vec{K}$-dependent boson quasiparticles of the dilute ferromagnet. [The latter can be determined only when the Green's function $\bar{G}_{g,g'}(E)$ has been solved for arbitrary separations $(\vec{g}-\vec{g}')$.] Nevertheless, since in a dense ferromagnet the proper elementary excitations coincide with the magnetic single-site spin waves, and because in the present study we have only been able to determine the magnetic single-site Green's function, it is interesting to examine the structure of these spin waves here.

Because $\tilde{J}_{(E)}$ is complex for all E except for $E=0$, therefore, from Eq. (7.24) it follows that the poles of the Green's function $G_{\vec{K}}(E)$ occur for complex values of E for all $\vec{K}$ values except that which self-consistently refers to the case $E=+i0$, i.e., for all K except for $K=0$. The important consequence of this circumstance is that even at absolute zero the spin waves are not exact normal modes (except for $K=0$) when the magnetic concentration m is different from unity. To illustrate this situation, in Fig. 2 we have plotted the real and the imaginary parts of the coherent exchange integral $J^{(E)}$, for several different concentrations, as a function of the energy. The examination of the imaginary part shows that in the long-wavelength limit, i.e., for small energy, the spin waves are good normal modes. The region of K for which this is true, however, continues to shrink as m decreases toward m_c. For $m=m_c+0$, only the $K=+0$ mode is undamped. The widths are seen to be largest at some intermediate value of energy which, say, corresponds to the length of wave vector equal to K_0. As m decreases toward m_c, K_0 also recedes towards $+0$. In the opposite limit, i.e., $K\to\Pi$, the contribution of the single-site spin waves to the density of states $\rho_m(\omega)$ decreases rapidly. (The displayed curves have been terminated at those values of the energy for which this contribution has become effectively zero.) Moreover, because the present calculation exactly preserves only the first few frequency moments of $\rho_m(\omega)$, its relative accuracy for larger values of ω can be expected to be poor. Both these considerations lead us to expect greatly increased errors in the results for $E_{\vec{K}}(\omega)$ and $\psi_{\vec{K}}(\omega)$ for large values of $\vec{K}$ and ω. As such, we should like to caution ourselves against taking the results that would follow from the given values of $\mathrm{Re}\,J^{(E)}$ and $\mathrm{Im}\,J^{(E)}$ for $\tilde{E}_{\vec{K}}(\tilde{E}_{\vec{K}})$ and $\psi_{\vec{K}}(\tilde{E}_{\vec{K}})$ too seriously beyond about $K\sim K_0$.

To conclude this section, we describe the magnetic site density of states $\rho_m(\omega)$ as a function of the concentration. In Fig. 3 we have plotted several of these curves. The general trend is clear. As the magnetic dilution increases, the center of gravity of the density of states shifts towards smaller frequencies. Moreover, the cutoff frequency ω_c beyond which $\rho_m(\omega)$ is effectively zero decreases with the increases of dilution.

IX. REMARKS ON ACCURACY

The truncated two-site neighborhood approximation studied in Secs. VII and VIII is subject to two different categories of errors. First there are the errors involved in the truncation of the neighborhood, i. e., from the full crystal size to a pair of neighboring atoms. Second, the computation used for the evaluation of $\tilde{J}_{(E)}$, and consequently the magnetic single-site spin-wave spectrum and the density of states $\rho_m(\omega)$, is also subject to numerical errors. The latter category of errors is, naturally, the easier one to estimate, and we shall describe it first.

The curves shown in Figs. 2 and 3 were computed by self-consistently requiring the absolute value of the left-hand side of Eq. (7.15) to be less than 10^{-5}. Moreover, the integral (8.1) was evaluated such that for the worst cases, i. e., small frequencies, the results for $\Gamma^{(0)}(E)$ were accurate to within about 2.5%.

By increasing the number of Newton-Raphson iteration cycles several fold, the absolute value of the left-hand side of Eq. (7.15) can be made sensibly vanishing, i. e., $< 10^{-9}$. Moreover, the accuracy of the numerical integration can be considerably increased by increasing the number of points used within the range τ, as well as the range τ itself, for the computation of the integral (8.1). [Here τ replaces the upper limit of ∞ as an approximation.] Incorporating these changes changed $J^{(E)}$ and $\rho_m(E)$ by about 2–2.5%. Because of the approximate nature of the present theory (due to

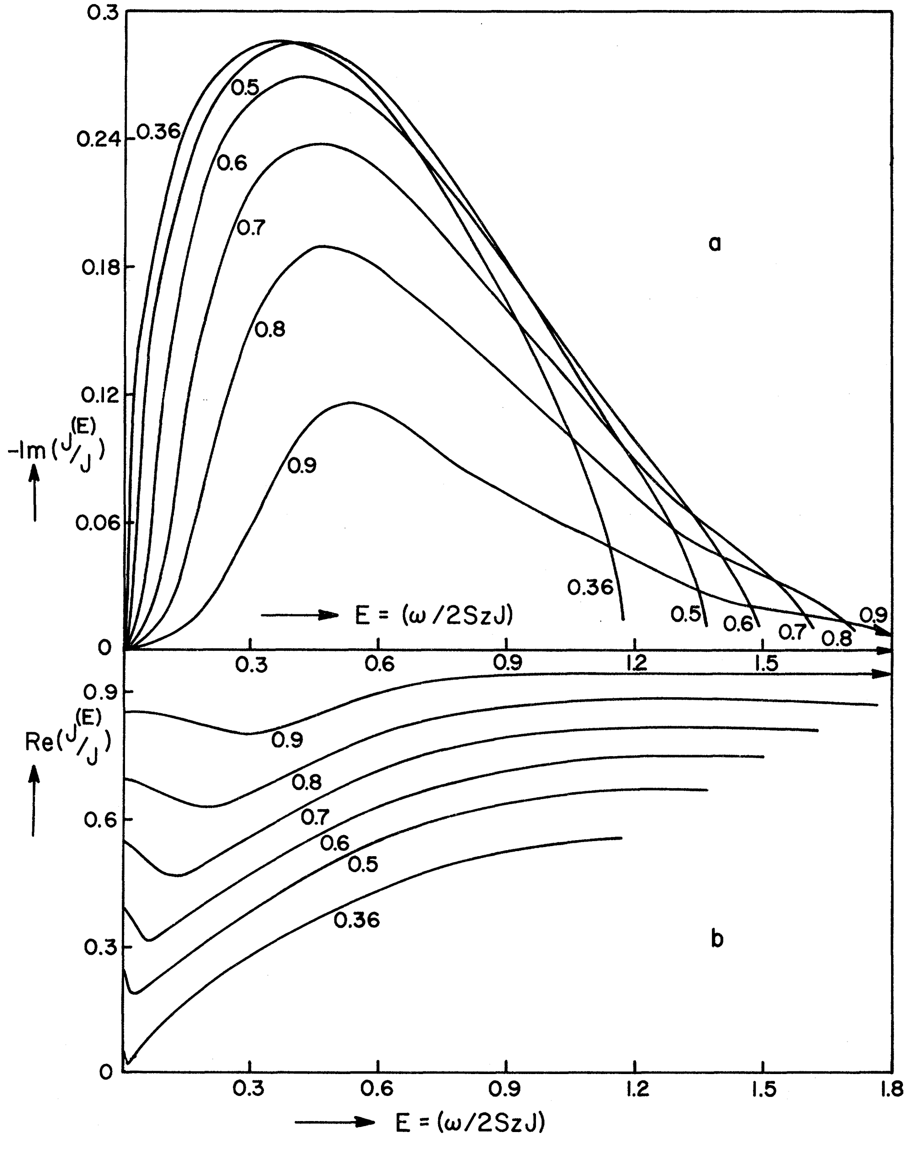

FIG. 2. (a) Negative imaginary part of the coherent exchange, $-\mathrm{Im}(J^{(E)}/J)$, as a function of the frequency for various values of the magnetic concentration in a simple-cubic lattice. The corresponding values of $\mathrm{Re}(J^{(E)}/J)$ are plotted in (b). Note that the absolute size of the imaginary part of the coherent exchange increases rapidly, relative to the real part, as the magnetic concentration m is reduced towards the critical concentration m_c. Note also that in this figure the notation $J^{(E)}$ is used to denote the nearest-neighbor coherent exchange integral $\tilde{J}_{(E)}$.

the drastic truncation of the Kohn neighborhood) this computational accuracy was not considered warranted. So the computational inaccuracy of the given numerical results in Figs. 2 and 3 can be assumed to be about 2.5%.

The estimate of the level of inaccuracy inherent in the truncation of the Kohn neighborhood to only two neighboring sites is much harder to obtain. Customarily, a reasonable gauge of the accuracy of self-consistent theories is considered to be provided by the number of frequency moments that are conserved exactly.[23,24] In our case, this means that we compare the exact results for the first several frequency moments of the density-of-states function $\rho_m(\omega)$ with those given by the approximate spectral function obtained from the Green's function $G_{0,0}(E)$ given in Eq. (7.4a).

The computation of the exact moments is carried out as follows: Assuming the presence of a magnetic atom on the site g, i.e., putting $\sigma_g = 1$, and writing

$$\langle [a_g(t),\ a_g^\dagger(t')]_- \rangle_m = \int \rho_m(\omega)\, e^{-i\omega(t-t')}\, d\omega\ , \qquad (9.1)$$

we have

$$\lim_{t=t'} \left\langle \left[\left(i \frac{d}{dt} \right)^\nu a_g(t),\ a_g^\dagger(t') \right]_- \right\rangle_m = \int_{-\infty}^{+\infty} \rho_m(\omega)\, \omega^\nu\, d\omega = M_\nu\ . \qquad (9.2)$$

[The suffix m under the right-hand pointed bracket denoting thermodynamic averaging indicates that the averaging in Eqs. (9.1) and (9.2) is conditional, being subject to the condition that a magnetic atom is known to be present on site g.]

The time derivatives of $a_g(t)$ are determined in the usual fashion by working out repeated commutators of a_g with $\mathcal{H}^c$ (subject to the condition $\sigma_g = 1$). Next, the commutator of these derivatives with $a_g^\dagger(t)$ is worked out. The only unknown quantity in the resultant expression is a correlation function of the form

$$\langle \sigma_{f_1} \cdots \sigma_{f_n} \rangle_m \equiv L_g(f_1, \ldots, f_n)\ . \qquad (9.3)$$

Because of the assumed spatial randomness of the substituted magnetic atoms, such correlation func-

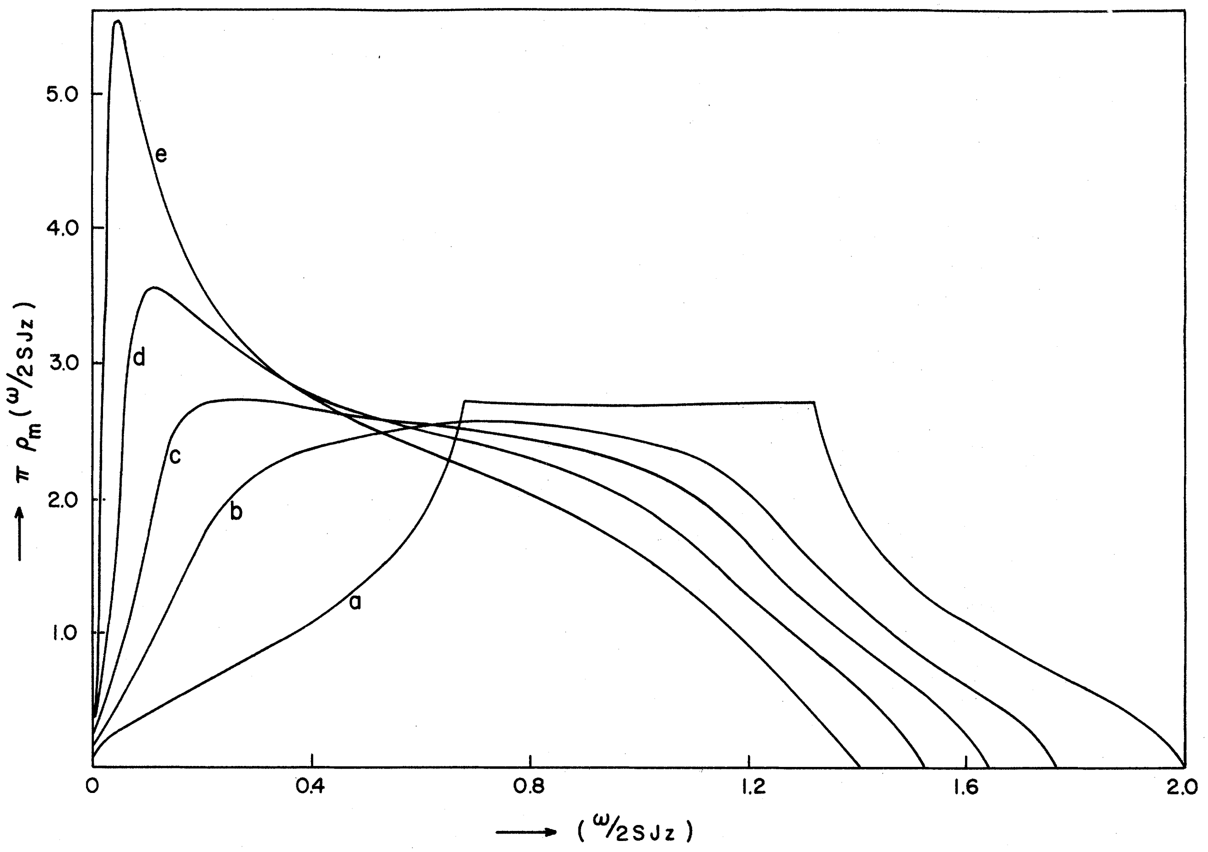

FIG. 3. For various magnetic concentrations m we show the magnetic-single-site density of states $\pi\rho_m(\omega/2Jsz)$. Curves a, b, c, d, and e, respectively, correspond to the magnetic concentration equal to 1.0, 0.8, 0.7, 0.6, and 0.5. For concentrations below 0.5, the curves peak much more rapidly, and at even lower frequencies. These results refer to a simple-cubic lattice.

tions are in principle straightforward to compute. In practice, their computation for $\nu \gg 1$ is quite tedious. Below we record the results for the first five moments calculated in this manner:

$$\Delta(0) = 1 \,, \tag{9.4a}$$

$$\Delta(1) = m \,, \tag{9.4b}$$

$$\Delta(2) = m^2 + (2m - m^2)/z \,, \tag{9.4c}$$

$$\Delta(3) = m^3 + (6m^2 - 3m^3)/z + [2m^3 - 6m^2 + 4m - m^2\phi(3)]/z^2 \,, \tag{9.4d}$$

$$\Delta(4) = m^4 + (12m^3 - 6m^4)/z + [11m^4 - 36m^3 + 25m^2 + m^3\phi(4) - 4m^3\phi(3)]/z^2 + [-6m^4 + 23m^3 - 23m^2 + 6m + 6\phi(3)(m^3 - m^2)]/z^3 \,, \tag{9.4e}$$

where

$$\Delta(\nu) = (2SJz)^{-\nu} M_\nu \,,$$

$$\phi(\nu) = z^2 \left(\frac{1}{N}\right) \sum_{\vec{K}} (\gamma_{\vec{K}})^\nu \,,$$

$$\begin{array}{lll} \phi(3) = 0 & \text{sc and bcc} & \\ = 4 \,, & \text{fcc} & \quad (9.4\text{f}) \\ \phi(4) = 2.5 & \text{sc} & \\ = 3.375 & \text{bcc} & \\ = 3.75 \,, & \text{fcc} \,. & \end{array}$$

The computation of the corresponding frequency moments within our nearest-neighbor-pair approximation of Sec. VII, is much simpler than the exact computation recorded above. Let us first recast Eqs. (7.14a) and (7.15) into a more convenient form:

$$\Gamma^{(0)}(E) = \frac{1}{N} \sum_{\vec{K}} \frac{1}{E - \eta(E)\,\alpha(\vec{K})} \,, \tag{9.5}$$

$$\eta(E) = \{mz - 2[1 - E\,\Gamma^{(0)}(E)]\} \times \{z - 2[1 - E\,\Gamma^{(0)}(E)]\}^{-1} \,, \tag{9.6}$$

$$\alpha(\vec{K}) = (2SJz)(1 - \gamma_{\vec{K}}) \,. \tag{9.7}$$

For $|E| \gg 1$ we can expand in inverse powers of E, i.e.,

$$\Gamma^{(0)}(E) = \sum_{\nu=0}^{\infty} (2SJz)^\nu \Delta(\nu)/(E)^{\nu+1} \,, \tag{9.8}$$

$$\eta(E) = \sum_{\nu=0}^{\infty} (2SJz)^\nu \eta^{(\nu)}/(E)^\nu \,. \tag{9.9}$$

Similarly, expanding Eq. (9.6) we get

$$E\,\Gamma^{(0)}(E) = \sum_{n=0}^{\infty} [\eta(E)]^n (2SJz)^n F(n)/(E)^n \,, \tag{9.10}$$

where

$$F(n) = \frac{1}{N} \sum_{\vec{K}} (1 - \gamma_{\vec{K}})^n \,. \tag{9.11}$$

Solving Eqs. (9.8)–(9.11) self-consistently in an order-by-order series expansion, we can readily calculate the frequency moments M_ν. The results for the moments M_0, M_1, and M_2 are found to be identical to the exact results given in Eqs. (9.4a)–(9.4f). For M_3 and M_4 the results are

$$\Delta(3) = m^3 + \frac{(6m^2 - 3m^3)}{z} + \frac{[2m^3 - 6m^2 + 4m - m^3\phi(3)]}{z^2} \,, \tag{9.12}$$

$$\Delta(4) = m^4 + \frac{(12m^3 - 6m^4)}{z} + \frac{[12m^4 - 36m^3 + 24m^2 + m^4\phi(4) - 4m^4\phi(3)]}{z^2} + \left(\frac{1}{z}\right)^3 [-4m^4 + 4m^3 - 8m^2 + 8m + 8\phi(3)(m^4 - m^2)] \,. \tag{9.13}$$

Because $\phi(3)$ is zero for the simple- and the body-centered-cubic lattices, for these lattices the present approximation exactly preserves the first four moments of the density of states, i.e., $M_0 - M_3$. The errors that arise in M_4 are seen to be of the order of $(1/z)^2$ of the dominant term.

Indeed, we have computed the two leading orders in $(1/z)$ for the exact and the approximate expressions for moments of arbitrary order, M_ν, and they coincide. [See Appendix.] Because the moments M_ν are dependent on the coefficients $\eta^{(0)} \cdots \eta^{(\nu-1)}$ [compare Eq. (9.9)], this in turn means that any arbitrary $\eta^{(\nu)}$ is determined exactly in the first two leading orders in z^{-1}, i.e., in an expansion of the form

$$\eta^{(\nu)} = \sum_{n=0}^{\infty} z^{-n} \eta_n^{(\nu)} \,, \tag{9.14}$$

the terms $\eta_0^{(\nu)}$ and $\eta_1^{(\nu)}$ are given exactly by the present approximation. Assuming the convergence of the expansion (9.9) for all E in the upper half of the complex energy plane, the reasonableness of the $1/z$ expansion given in Eq. (9.14) is very much a function of the size of z. For infinite z, the terms retained in Eq. (9.14) will continue to be a very accurate representation of $\eta^{(\nu)}$ even for large ν. However, for finite z, the convergence of the series (9.14) becomes extremely doubtful for $\nu \gtrsim z$. To our knowledge, this point seems not to have been emphasized in the literature dealing with $1/z$ expansions.

The foregoing considerations lead us to expect our results to be good in the limit $z \gg 1$. Indeed, for this case we can also give a heuristic interpretation for the magnitude of the critical concentration, i.e., $m_c = 2/z$. Let us consider an arbitrary site g in the lattice which happens to have a magnetic atom on it. Then around g there occurs a nearest-neighbor shell of z atoms with which this magnetic atom can interact via the exchange interaction, that is, whenever any of these atoms happens to be magnetic. Because the system is substitutionally random, on the average mz of these z atoms (surrounding the magnetic atom g) will be magnetic. Here we should remember that if z is large enough, the statistical fluctuation in the number of magnetic atoms surrounding g will be very small compared with mz, i. e., if $z \gg 1$, then $(z)^{1/2} \ll z$. Now, the highest value of mz for which the magnetic atoms will not form a three-dimensional network is equal to 2. To see this, imagine mz being equal to 2. Then for $z \to \infty$ two and only two of the nearest neighbors of g will be magnetic. Moreover, since each of these will have only two nearest neighbors which will be magnetic, and since one of them is g, with $m = 2/z$ the magnetic atoms will form linear chains. (Note these chains will not necessarily be straight.) And because linear Heisenberg spin chains cannot have magnetic long-range order,[27] the system will be nonmagnetic for $m < 2/z$. On the other hand, when $m > 2/z$, the magnetic atoms will begin to develop additional dimensionality. This dimensionality of the magnetic lattice will, however, not be two but rather it will be three. As three-dimensional Heisenberg spin lattices can sustain long-range magnetic order, we expect that our estimate for the critical magnetic concentration, being equal to $2/z$, is not unreasonable for $z \to \infty$.

Because of the extremely large statistical fluctuation in the number of magnetic nearest neighbors when z is as small as about 10, the foregoing argument cannot be used very convincingly for estimating the critical concentration for cubic lattices. Indeed, even the statement that the relative magnitude of the fluctuations will be proportional to $(z)^{-1/2}$ is no longer meaningful, for $z \sim 10$ is not a large number in the statistical sense. It is, however, reasonable to suspect that for such noninfinite z, the estimate of $2/z$ for the critical concentration indicates a certain lower bound. This conclusion follows from the reasoning that for finite z, fluctuations in the magnetic occupancy of the nearest-neighbor shell become large and consequently when $m \sim 2/z$, submacroscopic spatial regions, with average magnetic occupancy per nearest-neighbor shell much less or much more than mz, can exist. Under these circumstances the long interconnecting network of nearest-neighbor exchange coupled links necessary for the establishment of the long-range order breaks down even when m slightly exceeds the value $2/z$. Only when m is sufficiently larger than $2/z$ would the magnetic long-range order occur for finite z. As a very crude estimate for the amount by which the figure of $2/z$ may be an underestimate for m_c, the order of several times $(1/z^2)$ seems plausible. For the worst case in three dimensions, i. e., $z = 6$, this means that m_c may be expected to be $0.33 + \frac{1}{36} a_0$, where, as a convenient guess, we may take $1 < a_0 < 4$.

It is interesting next to compare our results for small impurity concentrations with those given by Murray,[12] Kaneyoshi,[18] and Edwards and Jones.[18] These authors agree on the result for the first-order correction to the spin-wave energy arising from the presence of a small concentration of nonmagnetic impurities, i. e.,

$$\tilde{J}_{(i0)}/J = 1 - b_0 (1 - m) + 0(1 - m)^2 , \quad 1 - m \ll m . \tag{9.15}$$

For large z the coefficient b_0 can be readily evaluated from the expressions given by these authors, and one finds

$$b_0 = 1 + 2/z + 0(1/z^2) . \tag{9.16}$$

This result is clearly reproduced by our theory.

For $z = 6$, these authors have estimated the coefficient b_0 to be ~ 1.532. Our theory, on the other hand, gives $b_0 = 1.5$ for the simple-cubic lattice. This discrepancy is expected because our theory is justified as a z^{-1} expansion scheme. However, while subject to the z^{-1} criterion, our theory may be expected to have some validity [unlike the perturbation schemes which expand in powers of the small parameter $(1 - m)$] even for large impurity concentrations, i. e., when $(1 - m) \sim m$.

In the light of the foregoing analysis, it is reasonable to expect that a procedure which makes use of a proper nearest-neighbor Kohn neighborhood would yield even more accurate results than the severely truncated two-atom neighborhood used in the present calculations. First, upon examining the structure of the frequency moments of the magnetic single-site density of states $\rho_m(\omega)$ (see for example the discussion given in the Appendix), we can reliably estimate that such a result for $\rho_m(\omega)$ will exactly conserve the moments M_0 to M_5. Moreover, because the neighborhood seems to include the influence of all the second-neighbor sites on the central site [see Figs. 1(a) and 1(b), for example], we might expect that it may well yield results which are exact to the first three dominant orders in $1/z$. In any event, this more detailed calculation is now being attempted and will be reported at a future date.[25]

Before concluding, it should be noted that a sensible application of a mean-field virtual-crystal-like approximation would have lead us to the result

$$\tilde{J}_{(E)} \xrightarrow[\text{mean field}]{} mJ \text{ for all } E . \tag{9.17}$$

This then would have given a density of states whose frequency moments would be exact only to the leading term in the z^{-1} expansion. In this sense, the present approximation may be looked at as being the first-order correction to the mean-field theory in a z^{-1} hierarchy. Therefore, the proper nearest-neighbor Kohn-neighborhood theory may be conjectured to be in the nature of a second-order correction.

X. SUMMARY AND COMMENTS

We have given a new, and seemingly very useful, formulation of the small-neighborhood cluster-averaged T-matrix approximation. The rationale and the philosophy behind this approximation procedure is in some ways similar to that expounded by Butler and Kohn[20] and others.[28] However, the details as well as the schematization are different.

Using a truncated version of this approximation procedure, we have discussed the magnetic single-site dynamics of a randomly dilute Heisenberg ferromagnet. Our results are, *a posteriori*, justified in an inverse z-expansion scheme and as such should have, for systems with $z \gg 1$, validity for larger values of the impurity concentration than the previous dynamical theories which rely on expansion schemes involving the impurity concentration as a small parameter. Nonetheless, because the accuracy of Kohn-like truncation schemes is in some sense dependent on the existence of a short intrinsic mean free path, our results for the exchange stiffness for extremely long-wavelength spin waves can be expected not to be totally reliable. We emphasize that the region of small frequencies is particularly vulnerable to inaccurate representation in the present theory—first, because of the inherent inaccuracy of the Kohn-like truncation for this region of long mean free paths and, second, because of the relatively large computational inaccuracy in this region.

In this regard the following comment may be made. Far from the critical regime, i.e., $m \sim 1$, the changes in the real part of the coherent exchange, $\tilde{J}_{(E)}$, with the frequency E are not large. Therefore, in this regime we may sensibly deal with finite frequencies which have appropriately short mean free paths and still extrapolate these results to the $E=0$ limit with a fair degree of accuracy. On the other hand, as m is decreased toward the critical regime, i.e., $m \sim m_c$, the damping at small but nonzero frequencies rises rapidly and at the same time the rate of change of the real part of $\tilde{J}_{(E)}$ becomes relatively more drastic. The net result of these competing processes, i.e., the increase in the damping helping out on the relative accuracy of the Kohn-like truncation procedure but the rapid change in $\tilde{J}_{(E)}$ for small E making the estimating of the Curie temperature more suspect, is that the small-neighborhood results are more reliable the farther from the critical concentration one is.

In order to give a somewhat more concrete description of the effects described above we have analyzed our numerical results for the simple-cubic system and we find that for long wavelengths the complex "normal-mode" dispersion relation may be approximately represented by the relation

$$\left(\frac{E_{\vec{K}}(m)}{E_{\vec{K}}(1)}\right)_{\vec{K}\ll 1} = \frac{zm-2}{z-2} + i\,\frac{D(m)K^{d}}{(m-m_c)^{n}} \;. \qquad (10.1)$$

In this expression the parameter $D(m)$ seems to have a structure which for narrow intervals of concentration m can be usefully represented as

$$D(m) \sim D_0(1-m) + O(1-m)^2 \,. \qquad (10.2)$$

Here the parameter D_0 is also concentration dependent. We feel that Eq. (10.2) could well be consistent with a power expansion of the form

$$D(m) = D_1(1-m) + D_2(1-m)^2 + \cdots \,, \qquad (10.3)$$

where D_1, D_2, etc., are independent of m.

In Eq. (10.1), the value of d seems to lie somewhere between 2.0 and 3.5, with possible inaccuracy of ±0.75. Similarly, the value of n in Eq. (10.1) is found to be somewhere near unity. Again the inaccuracy here may be about ±0.5.

As a crude guess, therefore, let us represent (10.1) as follows:

$$E_{\vec{K}}(m) = \left(\frac{m-m_c}{1-m_c}\right) 2JSK^2 + i\,C(m)\,(1-m)K^5/(m-m_c) \,, \qquad (10.4)$$

where $C(m)$ is concentration dependent through factors of the form $(1-m)$. Such a normal mode clearly changes character from a well-defined mode to a poorly defined phenomenon for wave vector $\vec{K}_c(m)$, where

$$\left(\frac{m-m_c}{1-m_c}\right) 2JSK_c^2 \sim C(m)\left(\frac{1-m}{m-m_c}\right) K_c^5 \,, \qquad (10.5)$$

that is, where

$$K_c(m) \sim (m-m_c)^{2/3\pm 1/3} / [C(m)\,(1-m)]^{1/3\pm 1/6} \,. \qquad (10.6)$$

As we recede from the $m=1$ limit, the transition from the well-defined mode to poorly defined mode occurs for shorter and shorter wave vectors. Indeed, if we define a characteristic length $l_c(m)$ such that $l_c(m)K_c(m) \sim 1$, then

$$l_c(m) \sim (m-m_c)^{-2/3}\, C^{1/3}(m)\,(1-m)^{1/3} \,. \qquad (10.7)$$

Therefore, when m is not close to unity and the term $(m-m_c)^{-2/3}$ dominates the behavior of the mean free path, we may expect that a suitable size of the Kohn neighborhood is going to have to increase by roughly the factor $(m-m_c)^{-2/3}$ in order to describe the dynamics of the randomly disordered system reasonably. This leads us to the most discouraging comment of this paper: For lattices with coordination z which is not $\gg 1$, we need to consider an infinitely large neighborhood to adequately describe the situation at the critical concentration!

Note added in proof. A more complete numerical analysis indicates that n in Eq. (10.1) is close to zero. Consequently, the exponent of $(m - m_c)$ in Eq. (10.7) is $-\frac{1}{3}$.

APPENDIX

In Sec. IX it was mentioned that the first two terms in the frequency moments of the magnetic single-site density of states, ordered in terms of a power expansion in z^{-1}, can be calculated. We shall show below that these terms are

$$\Delta(\nu) = (2SJz)^{-\nu} M_\nu = m^\nu + \nu(\nu - 1)(2m^{\nu-1} - m^\nu)/2z + R^{(\nu)}(z) . \tag{A1}$$

We recall from Eq. (9.2) that

$$M_\nu = \langle [(D)^\nu a_g(t),\ a_g^\dagger(t)]_- \rangle_m , \tag{A2}$$

where

$$D \equiv i \frac{d}{dt} , \tag{A3}$$

and where the conditional thermodynamic average on the right-hand side of (A2) is taken over all configurations subject to the condition that a magnetic atom is present at the site g, i.e., $\sigma_g = 1$.

Under the Hamiltonian (2.8), the right-hand side of Eq. (A2) can be determined by repeated application of the commutator,

$$D a_g = 2S \sum_f \sigma_f J(gf)(a_g - a_f) , \tag{A4}$$

and finally the use of the commutation rule (2.4). The resultant expressions contain a unique term $B(\nu)$ involving a spatial correlation function of ν spatial locations, i.e.,

$$B(\nu) = (2S)^\nu \sum_1 \cdots \sum_\nu J(g1) \cdots J(g\nu) \langle \sigma_1 \cdots \sigma_\nu \rangle_m . \tag{A5}$$

In this term none of the ν sites, labeled 1 to ν, can coincide with the site g. Because of the assumed randomness of the site locations of the magnetic lattice (except for the site g), the spatial correlation contributes a factor m^ν if all the ν locations $1, \ldots, \nu$ are different. However, because $(\sigma_i)^2 = \sigma_i$, we must subtract from this the situations where two or more of the sites are coincident. If we think in terms of a series expansion in inverse powers of z, then clearly the magnitude of the contribution of the terms involving coincidence of two or more sites to $B(\nu)$ will be at least an order z^{-1} smaller. If we are interested only in retaining the first two dominant powers of z, then $B(\nu)$ of Eq. (A5) is

$$B(\nu) = (2SJz)^\nu \left[m^\nu - \frac{m^\nu \nu(\nu - 1)}{2z} + m^{\nu-1} \frac{\nu(\nu - 1)}{2z} + O\left(\frac{1}{z^2}\right) \right] . \tag{A6}$$

Here the first term arises as a result of the completely unrestricted sum over sites $1, \ldots, \nu$, while the second term subtracts from the first the contribution arising from the coincident pairs that can be formed out of the ν sites. [Note that because in this term the sum is taken over only $(\nu - 1)$ free indices it is of order z^{-1} of the first term. Moreover, note that we do not subtract the contribution that is given by the equality of three or more of the ν sites because these terms are formally of lower order in z^{-1}.]

The third term on the right-hand side describes the situation that obtains when any two of the ν sites are coincident. Here, of course, we are adding a contribution $m^{\nu-1}$ because to the lowest order in z^{-1} the $(\nu - 1)$-site correlation gives $m^{\nu-1}$. All remaining terms on the right-hand side of (A6) are thus seen to be of order $(1/z^2)$ of the first term.

Next we consider the remaining $2^\nu - 1$ terms. First we notice that due to the occurrence of the spatial Kronecker δ function arising from the commutators in Eq. (A2), these terms contain only $(\nu - 1)$-site spatial correlations. (This is because $\sigma_g = 1$.) Second, we see that of these $2^\nu - 1$ terms, $2^{\nu-1}$ are negative and $2^{\nu-1} - 1$ are positive. All the negative terms contain closed polygons in a schematic representation where $J(f_1 f_2)$ is represented by a line connecting the neighboring sites f_1 and f_2. [For example, the lowest-order closed polygon is a triangle caused by the presence of a factor of the form $J(f_1 f_2) J(f_2 f_3) J(f_3 f_1)$, etc.] Because all terms containing closed polygons (note that these polygons may have any number of open branches), are at least an order $(1/z)$ lower than those which do not contain closed figures, to the order maintained in Eq. (A6) we can ignore these terms. [Note that the leading order in all such $2^\nu - 1$ terms can at most be of order $(z)^{\nu-1}$ because there are then only $(\nu - 1)$ free spatial sums involved.] Out of the $2^\nu - 1$ positive terms only $\frac{1}{2}[\nu(\nu - 1)]$ terms do not contain polygons (this being the number equal to the number of times only two of the ν sites coincide.) Therefore, their contribution in the leading order in $1/z$ is

$$C(\nu) = (2SJz)^\nu \left(\frac{m^{\nu-1}}{z} \frac{\nu(\nu - 1)}{2} + O(1/z)^2 \right) . \tag{A7}$$

Adding $B(\nu)$ and $C(\nu)$ and multiplying both sides $(2SJz)^{-\nu}$, we get the stated result given in Eq. (A1).

The remainder $R^{(\nu)}(z)$, in Eq. (A1), is clearly a complicated function of ν and z. Although, in an order by order expansion in z^{-1}, it must only contain terms proportional to z^{-n} with $n \geq 2$, we cannot in general claim that for all ν and z, $R^{(\nu)}(z)$ is small compared with the terms retained. Indeed, for finite z and infinite ν, the remainder will contain an infinite number of terms in each order of z^{-n} and therefore the cumulative effect of these terms could well vitiate the meaningfulness of z^{-1} expansion. For the opposite case, i.e., finite ν and infinite z, the situation is, however, simple. Here $R^{(\nu)}(z)$ must clearly be vanishingly small compared with the terms retained.

Let us next evaluate the equivalent leading terms in M_ν through the use of the approximate density of states $\rho_m(\omega)$ that the present approximation, using a two- (neighboring-) site Kohn neighborhood, gives. To this end we combine Eqs. (9.6)–(9.11) and write

$$\eta^{(0)}\left(\frac{z-2}{z}\right)+\frac{z-2}{2}\sum_{n=1}^{\infty}\frac{(2SJz)^n}{n}\,\eta^{(n)} + \left(\eta^{(0)}+\sum_{i=1}^{\infty}\frac{(2SJz)^i}{E^i}\,\eta^{(i)}\right)\left(1+\sum_{p=1}^{\infty}(2SJz)^p\,\frac{\Delta(p)}{E^p}\right) = \frac{zm}{2}+\sum_{j=1}^{\infty}\frac{(2SJz)^j}{E^j}\,\Delta(j)\ . \qquad \text{(A8)}$$

Comparing coefficients of E^{-n} on both sides of this equation we get $\eta^{(0)} = m$ and

$$\frac{z}{2}\,\eta^{(n)} + m\,\Delta(n) + \sum_{i=1}^{n-1}\eta^{(n-i)}\,\Delta(i) = \Delta(n)\ , \quad n \neq 0\ . \qquad \text{(A9)}$$

Because to the leading order in z^{-1}, $\Delta(n) \sim (m)^n$, according to Eq. (9.14) we have

$$\begin{aligned} \eta_0^{(n)} &= 0\ , && n \neq 0 \\ \eta_1^{(n)} &= -\frac{2}{z}\,(m^n - m^{n+1})\ , && n \neq 0\ . \end{aligned} \qquad \text{(A10)}$$

Next, combining Eqs. (9.8) and (9.10), noting that

$$F(n) = 1 + \frac{n(n-1)}{2z} + O\left(\frac{1}{z}\right)^2\ , \qquad \text{(A11)}$$

comparing coefficients of $(z/E)^\nu$ and $z^{-1}(z/E)^\nu$, and using Eq. (A10) we get the desired result for $\Delta(\nu)$. This result is identical to that given in (A1) to the leading two orders in z^{-1}. From our analysis of the fifth moment M_4, we expect that the remainder $R^{(\nu)}(z)$ given by the present approximation will not be identical to the exact one and as such we do not attempt to evaluate it here.

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[1] R. Brout, Phys. Rev. 115, 824 (1959).

[2] R. J. Elliott, J. Phys. Chem. Solids 16, 165 (1960).

[3] J. S. Smart, J. Phys. Chem. Solids 16, 169 (1960).

[4] P. W. Kasteleijn and J. van Kranendonk, Physica 2, 367 (1956).

[5] H. A. Brown and J. M. Luttinger, Phys. Rev. 100, 685 (1955).

[6] S. H. Charap, Phys. Rev. 126, 1393 (1962).

[7] R. J. Elliott and B. R. Heap, Proc. Roy. Soc. (London) A265, 264 (1962).

[8] B. R. Heap, Proc. Phys. Soc. (London) 82, 252 (1963).

[9] R. J. Elliott, B. R. Heap, D. J. Morgan, and G. S. Rushbrooke, Phys. Rev. Letters 5, 366 (1960).

[10] G. S. Rushbrooke and D. J. Morgan, Mol. Phys. 4, 1 (1961).

[11] D. J. Morgan and G. S. Rushbrooke, Mol. Phys. 6, 477 (1963).

[12] G. A. Murray, Proc. Phys. Soc. (London) 111, 89 (1966).

[13] H. Sato, A. Arrott, and R. Kikuchi, J. Phys. Chem. Solids 10, 19 (1959).

[14] C. Domb and M. F. Sykes, Phys. Rev. 122, 77 (1961).

[15] H. L. Frish, E. Sonnenblick, V. A. Vyssotsky, and J. M. Hammersley, Phys. Rev. 124, 1021 (1961); H. L. Frisch, J. M. Hammersley, and D. J. A. Welsh, ibid. 126, 949 (1962).

[16] M. F. Sykes and J. W. Essam, Phys. Rev. 133, A310 (1964).

[17] A. Brooks Harris (private communication).

[18] T. Kaneyoshi, Progr. Theoret. Phys. (Kyoto) 42, 477 (1969); S. F. Edwards and R. C. Jones, J. Phys. C 4, 2109 (1971).

[19] T. Matsubara and T. Kaneyoshi, Progr. Theoret. Phys. (Kyoto) 36, 695 (1967); T. Matsubara and F. Yonezawa, ibid. 34, 871 (1965); and 37, 1346 (1967); F. Yonezawa and T. Matsubara, ibid. 35, 759 (1966).

[20] W. H. Butler and W. Kohn, J. Res. Natl. Bur. Std. (U.S.) 74A, 443 (1970).

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

[22] W. Kohn and J. M. Luttinger, Phys. Rev. 108, 590 (1957).

[23] P. Soven, Phys. Rev. 156, 809 (1967).

[24] B. Velický, S. Kirkpatrick, and H. Ehrenreich, Phys. Rev. 175, 747 (1968).

[25] R. A. Tahir-Kheli, N. F. Berk, and D. Shazeer (unpublished).

[26] T. Wolfram and J. Callaway, Phys. Rev. 130, 2207 (1963).

[27] N. D. Mermin and H. Wagner, Phys. Rev. Letters 17, 1133 (1966).

[28] K. F. Freed and M. H. Cohen, Phys. Rev. B 3, 3400 (1971).