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Spatially Random Heisenberg Spins at Very Low Temperatures. II. Dilute Antiferromagnet with Nearest-Neighbor Substitutional Short-Range Order*

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A formulation for the computation of the single-site dynamics of a dilute Heisenberg antiferromagnet with isotropic nearest-neighbor exchange and arbitrary nearest-neighbor substitutional short-range order (SRO) is given. In the random limit, i.e., when the SRO is zero, the resultant density of states is shown to exactly conserve the first four frequency moments (of the spin-wave density of states) for all lattice structures which admit two nearest-neighbor interpenetrating sublattices and correctly give the leading two terms in the z^{-1} expansion for all frequency moments. In the presence of the SRO, the theory is shown to lead to correct results in the limit of perfect spatial correlation and to give cumulantlike decoupling results for the three- and higher-site substitutional correlations when these correlations are short ranged. The minimum relative concentration of the magnetic atoms for which the antiferromagnetic long-range order does not obtain is found to be $2Q/z$, where the renormalization factor Q depends both on the actual magnetic concentration m and the SRO parameter. For perfect clustering among the magnetic atoms, $Q=m$ and hence for all nonzero magnetic concentration the system orders, whereas for complete substitutional randomness, $Q=1$. Hence antiferromagnetic long-range order obtains only when the relative magnetic concentration is higher than $2/z$.

I. INTRODUCTION

The study of dilute antiferromagnets has received a great impetus by the recent experiments by Breed *et al.*[1] on the mixed system $\mathrm{KMn}_{(m)}\mathrm{Mg}_{(1-m)}\mathrm{F}_3$. Analyzing it as a randomly dilute isotropic-exchange Heisenberg antiferromagnet with simple-cubic (magnetic) lattice structure, Jones and Edwards[2] and Osborne[3] have, respectively, predicted magnetic critical concentrations of 0.34 and 0.296. These predictions are fairly close to the experimental result of 0.31 measured by Breed *et al.*[1]

The philosophy behind the theories of Jones and Edwards[2] and of Osborne[3] is in some sense similar to that of perturbation theory. For instance, Jones and Edwards[2] sum a certain subset of the terms contributing in the linear power of the quantity $(1-m)$ to the spin-wave mass operator. Similarly, Osborne,[3] after generalizing the Green's-function perturbation-theoretic formalism of Kaneyoshi[4] used in the study of dilute ferromagnet to the antiferromagnetic case, computes his results consistently only to the simplest first-order perturbation theory.

In contrast to these theories, the rationale behind the present work is not perturbation theoretic. Rather, interpreting the philosophy of the Butler-Kohn[5] and Freed-Cohen[6] works somewhat loosely, we have recently argued[7] that the dynamical properties of a sufficiently random many-body system will be well approximated by the situation obtaining in typical small neighborhoods. In particular, it was argued that for any given frequency ω, if the size of the typical neighborhood is characterized by a length L, then the errors involved in equating the dynamical properties of the thermodynamic system with those of the neighborhood of dimension L would become exponentially small with the ratio $L/\lambda_0(\omega)$. Here $\lambda_0(\omega)$ is some typical mean free path of the physical system for the frequency ω.

In three dimensions, the smallest proper neighborhood which includes the effects caused by random interparticle scattering potentials was shown[7] to consist of 25 atoms (for a simple-cubic lattice, that is). For a binary system (such as a dilute antiferromagnet), this entails a maximum of $(2)^{25}$ configurations, which record the different occupations of such a neighborhood. In spite of the fact that the symmetry of the problem cuts down this number by several orders of magnitude, the resultant calculation is far from simple.[8] To keep the calculation tractable, we work with an exceedingly simple, drastically truncated, two-site neighborhood.[7] In spite of its extreme simplicity, we are able to extract fairly accurate results from such a procedure. The reason behind such an optimum use of this small-neighborhood approximation lies in our study of the conditional thermodynamic averages rather than the full thermodynamic averages.

Our results for the magnetic single-site density of states for the substitutionally random case exactly preserve the first four frequency moments of the density of states and are asymptotically exact in the mean-field limit, i.e., $z\rightarrow\infty$. Indeed, as for the dilute ferromagnet, these results are dynamically correct to the two leading orders in the z^{-1} expansion, as is shown by their preservation of all frequency moments of the magnetic single-site density of states to this order.

In the above, the exact results imply the results obtained in the spin-wave approximation, which assumes the quasiclassical structure for the concentrated antiferromagnetic ground state. As is well known, the spin-wave representation of the ground state is not exact and it underestimates the zero-point quantum fluctuations. Nevertheless, for the purposes of the present study, we assume the correctness of the spin-wave theory in the concentrated limit.

In Sec. II, a formulation of the completely random dilute antiferromagnet is given. Here the quasiclassical nature of the spin-wave representation of the ground state is made precise by a comparative discussion of the partial conditional averages and the full thermodynamic averages. The mathematical preliminaries are introduced and the T-matrix equations, in the presence of a nonunitary metric Λ, are derived in Sec. III. The properties of the coherent Green's function are analyzed in Sec. IV, while the truncated two-site-neighborhood T-matrix ansatz is introduced in Sec. V.

Section VI deals with a study of the frequency moments of the density of states for the spatially random case. Here the exactness of the first four frequency moments and the asymptotic exactness (to the two leading orders in z^{-1}) of all the frequency moments of the magnetic single-site density of states are demonstrated.

In the concluding sections (Secs. VII and VIII) the results of this paper are generalized to the case with nearest-neighbor short-range order (SRO) and a discussion of the noninfinite z case is given. It is shown how the presence of the SRO modifies the system Néel temperature for any given magnetic concentration. Moreover, the expression for the minimum concentration m_0 for which the system does not develop antiferromagnetic long-range order (LRO) is obtained as a function of the SRO, and it is shown how the dependence of m_0 on the SRO is entirely in accord with a heuristic qualitative picture. Next, by an examination of the many-atom spatial correlation function (that occurs in the expression for an arbitrary-order moment), it is shown that, if a cumulant-like decoupling of the many-atom clustering effects into those of neighboring pairs occurs, then these results are exact in the leading order in z^{-1}. Also, the first three moments are conserved exactly. Consequently, for nonzero SRO, the present approximation can be considered to be a reasonable interpolation scheme, because it gives exact results in the limit of perfect SRO, very reasonable results for zero SRO, and adequate results in between. Finally, in Sec. VIII, the case of noninfinite z is discussed and it is emphasized that for this case the present results can be expected to give a certain lower bound for the critical concentration.

II. FORMULATION

We consider a randomly dilute Heisenberg antiferromagnet with isotropic nearest-neighbor exchange interaction and vanishingly small staggered external fields. The directions of the staggered fields are taken to be along the positive z direction for the A sublattice, and along the negative z direction for the B sublattice.

As mentioned earlier, even in the concentrated limit, where all the N magnetic lattice sites are occupied, the ground state of the antiferromagnet is not exactly known. Because our primary interest here is in the dynamics of randomness, we make the usual quasiclassical assumption that for the concentrated case the ground state is described by a pair of up-down interpenetrating sublattices, with zero-point sublattice magnetization fluctuations which are adequately given by noninteracting spin-wave approximation. At $T=0$, a single spin-flip excitation is an exact eigenmode in this approximation, and it has a Bloch-like propagating solution which in the limit of long wavelength, i.e., $K \ll 1$, has the energy

$$E_K \approx DK \ . \tag{2.1a}$$

The parameter D depends upon the exchange interaction J, the magnitude of the spin S (we use Dirac units whereby $\hbar=1$), and the coordination number z of the lattice,

$$D = 2SzJ/\sqrt{3} \ . \tag{2.1b}$$

(We use the nearest-neighbor separation as the unit of length and consider only those lattices which are three dimensional and which do not have nearest-neighbor triangles. Moreover, throughout this paper we shall keep the magnitude of z as arbitrary, subject to the restriction $z \geqslant 6$.) As the temperature rises, the sublattice magnetization and the elementary excitation energy decrease and the modes develop finite lifetimes. When the system temperature reaches a critical temperature T_c, where $k_B T_c \sim \frac{1}{2}D(S+1)$, the system undergoes a transition to a phase with no magnetic long-range order.

As the magnetic occupation is reduced from this concentrated (i.e., fully occupied) limit by the substitution of nonmagnetic impurities in place of some of the magnetic atoms, the description of the magnetic state becomes even more complicated. Now, even within the aforementioned quasiclassical approximation, the ground state cannot be determined exactly for it depends upon the precise configuration in which the Nm magnetic and

$N(1-m)$ nonmagnetic atoms are distributed over the N available sites. A consequence of this randomness is that spins belonging to the A sublattice are no longer guaranteed to always have only B sublattice spins as their nearest neighbors and vice versa. In spite of the randomness, however, the dictate of the quasiclassical assumption regarding the antiferromagnetic ground state is the complete symmetry of the up-down sublattices. This means that, given an even number of magnetic atoms, the up-down sublattices will be equally populated.

The object of the present study is to investigate the structure of the zero-temperature long-wavelength spin-wave excitation $E_{\vec{K}}$ as a function of the magnetic concentration m. Assuming that the renormalization of $E_{\vec{K}}$, with the concentration m, has the same form as the renormalization of the critical temperature T_c, we will compute $T_c(m)/T_c(1)$. Moreover, the lowest concentration m_c for which the long-wavelength spin wave $E_{\vec{K}}$ has positive or zero energy will be computed. This concentration, for which the antiferromagnetic LRO does not obtain if the corresponding $D=0$, will be called the critical concentration m_0.

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, being independent of the magnetic concentration, is strictly a two-body object, we can write the system Hamiltonian as

$$\mathcal{H}^c(t) = -\sum_{f,\lambda} \mu^\lambda S_f^z(\lambda,t)\,\sigma_f^\lambda + 2{\sum_{f_1,f_2}}' J^{\lambda,-\lambda}(f_1,f_2) \times \sigma_{f_1}^\lambda \sigma_{f_2}^{-\lambda}\, \vec{S}_{f_1}(\lambda,t)\cdot\vec{S}_{f_2}(-\lambda,t) = \mathcal{H}^c \ . \tag{2.2}$$

Here the index λ denotes the "up" and the "down" magnetic sublattices according to whether $\lambda = A$ or $\lambda = B$. For notational convenience we define

$$A \equiv -B \ . \tag{2.3a}$$

As stated earlier we shall consider the limiting case for which the staggered Zeeman fields are vanishing, i.e.,

$$\mu^\lambda = -\mu^{-\lambda} = +0 \ . \tag{2.3b}$$

The magnetic occupation operators σ_f^λ are defined as

$$\begin{aligned} \sigma_f^\lambda &= 1 \quad \text{if the site } f \text{ is occupied by a spin belonging to the } \lambda \text{ sublattice} \\ &= 0 \quad \text{otherwise.} \end{aligned} \tag{2.4}$$

The first sum on the right-hand side of Eq. (2.2) is over all N sites, f, and over $\lambda = A$ and B. The second sum, denoted with a prime, is, however, a restricted sum such that it includes only distinct pairs (f_1, f_2) of sites and thus excludes any double counting of the same pairs. The exchange integral $J^{\lambda,-\lambda}(f_1,f_2)$ is a c-number function with the property

$$\begin{aligned} J^{\lambda,-\lambda}(f_1,f_2) &= J \quad \text{if } f_1 \text{ and } f_2 \text{ are nearest neighbors} \\ &= 0 \quad \text{otherwise.} \end{aligned} \tag{2.5a}$$

It should be recognized that, insofar as the Hamiltonian $\mathcal{H}^c$ depends explicitly upon the manner in which the mN magnetic atoms have been distributed over the N sites of the lattice, it is configuration dependent. Finally, the time dependence is in the Heisenberg notation with respect to the Hamiltonian $\mathcal{H}^c$, i.e.,

$$S_g^\alpha(\lambda,t) = e^{i\mathcal{H}^c t} S_g^\alpha(\lambda,0)\, e^{-i\mathcal{H}^c t}, \quad \alpha = x, y, z. \tag{2.5b}$$

We shall assume that the manner of occupation of the N sites is completely random. So, in thermal equilibrium, any given site has a probability m of being occupied by a magnetic atom and a probability $1-m$ of being occupied by a nonmagnetic atom.

To make this idea more precise, it is necessary to first define the concept of a configuration-dependent thermal-averaging process, i.e.,

$$\langle\Omega\rangle_c \equiv \mathrm{Tr}_c(e^{-\beta\mathcal{H}^c}\Omega)/\mathrm{Tr}(e^{-\beta\mathcal{H}^c}) \ . \tag{2.6}$$

The trace in Eq. (2.6) is taken only over those states which are consistent with the given configuration in which the Nm magnetic and $N(1-m)$ nonmagnetic atoms have been placed over the N sites. Next, we define the full thermal average

$$\langle\Omega\rangle \equiv \sum_{\{c\}} P(c)\langle\Omega\rangle_c \ . \tag{2.7}$$

Here $P(c)$ is the appropriate probability of the configuration over which the average $\langle\Omega\rangle_c$ has been determined and the sum $\{c\}$ is over all those configurations which are consistent with the total number of magnetic and nonmagnetic atoms being equal to Nm and $N(1-m)$, respectively. A simple consequence of this is that the thermodynamic averages of operators σ_i^λ and $\sigma_i^{-\lambda}$ are equal to $\frac{1}{2}m$ each, i.e., because

$$\langle\sigma_i^\lambda + \sigma_i^{-\lambda}\rangle = m \ ; \tag{2.8a}$$

therefore

$$\langle\sigma_i^\lambda\rangle = \langle\sigma_i^{-\lambda}\rangle = \tfrac{1}{2}m \ . \tag{2.8b}$$

(The equality of these averages is an essential part of our quasiclassical assumption about the antiferromagnetic ground state.)

Although the configurational and the unrestricted thermodynamic averages mentioned in the foregoing are of interest, of even greater importance to us is the process of conditional averaging. The simplest examples of this type of averaging are the following one- and two-body occupation correlation functions, e.g., $\langle\sigma_0^\lambda\rangle_{(0,\lambda)}$ and $J^{\lambda,-\lambda}(0,1)\times\langle\sigma_0^\lambda\sigma_1^{-\lambda}\rangle_{(0,\lambda)}$. The presence of the subscript $(0,\lambda)$ in an average specifies that the position 0 is known to be occupied by a magnetic spin which at $T=0$ can (in the quasiclassical sense made explicit earlier) be identified as belonging to the λ magnetic sublattice. Such averages are obtained by making use of the relation (2.7) after choosing, and summing over, only those configurations $\{c\}$ which are consistent with the condition that the site 0 is always occupied by a spin λ. Clearly then

$$\langle\sigma_0^\lambda\rangle_{(0,\lambda)}=1\ . \tag{2.9a}$$

Moreover, as a consequence of the quasiclassical assumption about the antiferromagnetic ground state, we have

$$\begin{aligned}J^{\lambda,-\lambda}(0,1)\langle\sigma_0^\lambda\sigma_1^{-\lambda}\rangle_{(0,\lambda)}&=J^{\lambda,-\lambda}(0,1)\langle\sigma_1^{-\lambda}\rangle_{(0,\lambda)}\\&=J^{\lambda,-\lambda}(0,1)m\ .\end{aligned} \tag{2.9b}$$

Note that the occurrence of the factor m on the right-hand side of Eq. (2.9b) is central to the quasiclassical assumption regarding the ground state. It says that since the site 1 is the nearest neighbor of site 0 [if it were not, $J^{\lambda,-\lambda}(0,1)$ would be vanishing], and since the site 0 is known to be occupied by a spin belonging to sublattice λ, the site 1 will have the probability m of being occupied by a spin of variety $-\lambda$, while it will have zero probability of being occupied by a spin belonging to sublattice $+\lambda$, and, of course, it will have probability $(1-m)$ of being occupied by a nonmagnetic atom. In the limit that all sites are magnetic, i.e., $m=1$, this assumption clearly reduces to the usual spin-wave-theory assumption for the concentrated ground state. [The contrast between the results of the full thermodynamic averages given in Eqs. (2.8a) and (2.8b) and the conditional averages given above in Eqs. (2.9a) and (2.9b) should be noted.]

III. MATHEMATICAL PRELIMINARIES

Let us introduce a temperature- and configuration-dependent double-time retarded Green's function with explicit reference to two spatial locations g and g' in the lattice:

$$\begin{aligned}G_{g,g'}^{\lambda,\lambda';c}(t,t')&=-2\pi i\,\Theta(t-t')\langle[S_g^+(\lambda,t);S_{g'}^-(\lambda',t)]_-\rangle_c\\&\equiv\langle\langle S_g^+(\lambda,t);S_{g'}^-(\lambda',t')\rangle\rangle_c\ .\end{aligned} \tag{3.1}$$

The function $\Theta(t-t')$ is the usual Heaviside step function. [See Eq. I(3.2). Here and henceforth we shall refer to the equations of Ref. 7 by putting the prefix I in front of the equation number.] The thermal average on the right-hand side of Eq. (3.1) is the configuration-dependent average defined in Eq. (2.6) and the notation $[X,Y]_-$ stands for the commutor $XY-YX$. The spin-flipping operators $S_g^\pm(\lambda,t)$, etc., are defined in the usual fashion.

The equation of motion of such a Green's function is easily determined by using the usual spin commutation relations (note that spin operators belonging to different sublattices commute) and the Hamiltonian defined in Eqs. (2.2)–(2.5a). Thus

$$\begin{aligned}i\frac{d}{dt}G_{g,g'}^{\lambda,\lambda';c}(t,t')&=2S_\lambda\,\delta_{g,g'}\,\delta_{\lambda,\lambda'}\,2\pi\delta(t-t')\\&\quad+2\sum_f J^{\lambda,-\lambda}(g,f)\,\sigma_g^\lambda\sigma_f^{-\lambda}\langle\langle S_g^z(\lambda,t)\,S_f^+(-\lambda,t)\,;S_{g'}^-(\lambda',t')\rangle\rangle_c\\&\quad-2\sum_j J^{\lambda,-\lambda}(g,j)\,\sigma_g^\lambda\sigma_j^{-\lambda}\langle\langle S_j^z(-\lambda,t)\,S_g^+(\lambda,t)\,;\ S_{g'}^-(\lambda',t')\rangle\rangle_c\ .\end{aligned} \tag{3.2}$$

In addition to the quasiclassical approximation relating to the antiferromagnetic ground state, henceforth we shall assume also that the dynamical operators $S_g^z(\lambda,t)$ and $S_j^z(-\lambda,t)$ can be replaced by their full thermal averages S_λ and $S_{-\lambda}$, i.e.,

$$S_g^z(\lambda,t)\sim\langle S_g^z(\lambda,t)\rangle=S_\lambda=-S_{-\lambda}=-\langle S_j^z(-\lambda,t)\rangle\approx-S_j^z(-\lambda,t). \tag{3.3}$$

This assumption is equivalent to the usual random-phase-approximation (RPA) decoupling assumption. As in the ferromagnet, if a suitable transformation is introduced, then the present decoupling procedure can readily be made to yield a set of relations identical to those following from the use of noninteracting $T=0$ spin-wave theory. For convenience, we shall defer a discussion of this point till later (see Sec. VI). Thus, Eq. (3.2) reduces to

$$\begin{aligned}i\frac{d}{dt}-2S_\lambda\Big[\sum_j J^{\lambda,-\lambda}(g,j)\,\sigma_g^\lambda\sigma_j^{-\lambda}\Big]\ G_{g,g'}^{\lambda,\lambda';c}(t,t')\\=2S_\lambda\,\delta_{g,g'}\,\delta_{\lambda,\lambda'}\,2\pi\,\delta(t-t')\\+2S_\lambda\sum_f J^{\lambda,-\lambda}(g,f)\,\sigma_g^\lambda\sigma_f^{-\lambda}\,G_{f,g'}^{-\lambda,\lambda';c}(t,t')\ .\end{aligned} \tag{3.4}$$

Because the system is not subject to external time-varying fields [i.e., $\mathcal{H}^c(t)$ and $\mathcal{H}^c(t')$ commute]

$$\begin{aligned}G_{g,g'}^{\lambda,\lambda';c}(t,t')&=G_{g,g'}^{\lambda,\lambda';c}(t-t')\\&=\int_{-\infty}^{+\infty}G_{g,g'}^{\lambda,\lambda';c}(E)\,e^{-iE(t-t')}dE\ .\end{aligned} \tag{3.5}$$

Exploiting this temporal translational symmetry, we can rewrite Eq. (3.4) in the following form:

$$EG^{\lambda,\lambda';c}_{g,g'}(E)-2S_\lambda\{\sum_f J^{(E)}(gf)\,G^{-\lambda,\lambda';c}_{f,g'}(E)+[\sum_j J^{(E)}(gj)]\,G^{\lambda,\lambda';c}_{g,g'}(E)\}$$

$$=2S_\lambda\,\delta_{g,g'}\,\delta_{\lambda,\lambda'}+\sum_{f,\nu}V^{\lambda,\nu;c}_{g,f}(E)G^{\nu,\lambda';c}_{f,g}(E), \qquad (3.6\text{a})$$

where

$$V^{\lambda,\nu;c}_{g,f}(E)=2S_\lambda[J^{\lambda,\lambda}(g,f)\,\sigma^\lambda_g\sigma^{-\lambda}_f-J^{(E)}(gf)]\,\delta_{\nu,-\lambda}+\delta_{g,f}\,\delta_{\lambda,\nu}\,2S_\lambda\{\sum_j[J^{\lambda,-\lambda}(g,j)\,\sigma^\lambda_g\sigma^{-\lambda}_j-J^{(E)}(gj)]\}\;. \qquad (3.6\text{b})$$

[These equations are obtained by first Fourier transforming Eq. (3.4) according to (3.5), and then by subtracting the second term on the left-hand side of Eq. (3.6a) from both sides of the resulting equation. Note that the function $J^{(E)}(f_1f_2)$ is as yet arbitrary. Its choice, as in Ref. 7, will be motivated by a physical criterion which will be described in a later section.] Introducing the notation

$$[\Gamma^{-1}(E)]^{\lambda,\nu}_{g,f}=E\,\delta_{\lambda,\nu}\,\delta_{g,f}-2S_\lambda[\delta_{\lambda,-\nu}J^{(E)}(gf)+\delta_{g,f}\,\delta_{\lambda,\nu}\sum_j J^{(E)}(gj)]\;, \qquad (3.7)$$

we can rewrite Eq. (3.6a) as

$$\sum_{f,\nu}[\Gamma^{-1}(E)]^{\lambda,\nu}_{g,f}\,G^{\nu,\lambda';c}_{f,g'}(E)=2S_\lambda\,\delta_{g,g'}\delta_{\lambda,\lambda'}+\sum_{f,\nu}V^{\lambda,\nu;c}_{g,f}(E)\,G^{\nu,\lambda';c}_{f,g'}(E)\;. \qquad (3.8)$$

Let us now introduce a matrix notation such that

$$\mathrm{G}^c(E)\equiv[G^{\nu,\nu';c}_{i,j}(E)]\;, \qquad (3.9\text{a})$$

$$\mathrm{V}^c(E)\equiv[V^{\nu,\nu';c}_{i,j}(E)]\;, \qquad (3.9\text{b})$$

$$\Gamma^{-1}(E)\equiv\{[\Gamma^{-1}(E)]^{\nu,\nu'}_{i,j}\}\;, \qquad (3.9\text{c})$$

$$\Lambda\equiv(2S_\nu\,\delta_{\nu,\nu'}\delta_{i,j})\;. \qquad (3.9\text{d})$$

These matrices are $(N\times 2)^2$ matrices such that their (initial and final) state indices refer to lattice positions along with sublattice indices ν and ν', i.e., A or B. Using this formal notation, Eq. (3.8) is written in the following succinct form:

$$\Gamma^{-1}(E)\,G^c(E)=\Lambda+V^c(E)\,G^c(E)\;. \qquad (3.10)$$

Premultiplying both sides of this by $\Gamma(E)$, where we define $\Gamma(E)$ by the relation

$$\Gamma(E)\,\Gamma^{-1}(E)=1=\Gamma^{-1}(E)\,\Gamma(E)\;, \qquad (3.11)$$

we get

$$G^c(E)=\Gamma(E)\,\Lambda+\Gamma(E)\,V^c(E)\,G^c(E)\;. \qquad (3.12)$$

This is the central equation of this section. However, it is convenient to transform it further as follows: Let us define a T matrix such that

$$V^c(E)\,G^c(E)=T^c(E)\,\Gamma(E)\,\Lambda\;. \qquad (3.13)$$

Then Eq. (3.12) would become

$$G^c(E)=\Gamma(E)\,\Lambda+\Gamma(E)\,T^c(E)\,\Gamma(E)\;. \qquad (3.14)$$

Moreover, this relation, when both of its sides are premultiplied by $V(E)$, i.e.,

$$V^c(E)\,G^c(E)=V^c(E)\,\Gamma(E)\,\Lambda+V^c(E)\,\Gamma(E)\,T^c(E)\Gamma(E)\Lambda\;, \qquad (3.15)$$

implies the following [if we use (3.13) for its left-hand side]:

$$[-T^c(E)+V^c(E)+V^c(E)\Gamma(E)\,T^c(E)]\,\Gamma(E)\,\Lambda=0. \qquad (3.16)$$

As $\Gamma(E)\Lambda$ is arbitrary (depending, as it does, on an arbitrary function), we get

$$T^c(E)=V^c(E)+V^c(E)\,\Gamma(E)\,T^c(E)\;. \qquad (3.17)$$

[Matrix elements of $T^c(E)$ will be denoted as $T^{\lambda,\lambda';c}_{i,j}(E)$.]

The central equation, (3.12), has thus been reduced to the two relations (3.14) and (3.17).

IV. MATRIX $\Gamma(E)$

In Sec. III we introduced a matrix $\Gamma^{-1}(E)$ by defining its matrix elements in the product space of lattice positions and the sublattice indices according to Eq. (3.7). Because this matrix depends upon the as yet arbitrary parameter $J^{(E)}(ij)$, its properties are similarly arbitrary.

In this section it is convenient to discuss the properties of the matrix $\Gamma(E)$ subject to the condition that the parameter $J^{(E)}(ij)$ satisfies the following requirement:

$$J^{(E)}(ij)=J^{(E)}\quad\text{if } i \text{ and } j \text{ are nearest neighbors}$$
$$=0\quad\text{otherwise.} \qquad (4.1)$$

The physical relevance of the choice of (4.1) will be made clear in Sec. V. For the present it only need be said that within a certain physically meaningful ansatz, an appropriate choice for $J^{(E)}$ can be made for which the conditionally averaged thermodynamic Green's-function matrix element $G^{\lambda;\lambda}_{0,0}(E)$ is equal to $\Gamma^{\lambda;\lambda}_{0,0}(E)$ to a good approximation. Here

$$G^{\lambda;\lambda}_{0,0}(E)=\sum_{\{c,\sigma^\lambda_0\}}P^\lambda_0(c)\,G^{\lambda;\lambda}_{0,0}(E)\;, \qquad (4.2)$$

$P^\lambda_0(c)$ is the probability of the occurrence of a configuration c which is consistent with the occupation of the site 0 by a magnetic atom belonging to the magnetic sublattice λ, and $\{c,\sigma^\lambda_0\}$ is the total set of such configurations that are possible given the existence of Nm magnetic atoms and $N(1-m)$ nonmagnetic ones.

Expanding the matrix element of $\Gamma^{-1}(E)\Gamma(E)$ as

$$[\Gamma^{-1}(E)\Gamma(E)]^{\lambda,\lambda}_{g,p}=\sum_{f,\nu}[\Gamma^{-1}(E)]^{\lambda,\nu}_{g,f}\,[\Gamma(E)]^{\nu,\lambda}_{f,p}\;, \qquad (4.3\text{a})$$

noting the identity

$$(1)^{\lambda,\lambda'}_{g,p}=\delta_{\lambda,\lambda'}\delta_{g,p}\ , \tag{4.3b}$$

using the expression for $[\Gamma^{-1}(E)]^{\lambda,\nu}_{g,f}$ given in Eq. (3.7), and equating (4.3a) with (4.3b) for $\lambda=\lambda'$ according to Eq. (3.11), we find

$$E\Gamma^{\lambda,\lambda}_{g,p}(E)=\delta_{g,p}+2S_\lambda\Big[\sum_j J^{(E)}(gj)\Big]\Gamma^{\lambda,\lambda}_{g,p}(E)+2S_\lambda\sum_f J^{(E)}(gf)\Gamma^{-\lambda,\lambda}_{f,p}(E)\ . \tag{4.4a}$$

Similarly, we also find

$$E\Gamma^{-\lambda,\lambda}_{l,p}(E)=-2S_\lambda\sum_f J^{(E)}(lf)\,\Gamma^{\lambda,\lambda}_{f,p}(E)-2S_\lambda\Big[\sum_j J^{(E)}(lj)\Big]\Gamma^{-\lambda,\lambda}_{l,p}(E)\ . \tag{4.4b}$$

Because $\Gamma^{\lambda,\nu}_{i,j}(E)$ depends upon the vector separation $(\vec{\mathrm{i}}-\vec{\mathrm{j}})$, we can use the Fourier transformation

$$\Gamma^{\lambda,\nu}_{i,j}(E)=\frac{1}{N}\sum_{\vec{K}}e^{i\vec{K}\cdot(\vec{\mathrm{i}}-\vec{\mathrm{j}})}\Gamma^{\lambda,\nu}_{\vec{K}}(E)\ , \tag{4.5a}$$

along with

$$J^{(E)}(ij)=\frac{zJ^{(E)}}{N}\sum_{\vec{K}}e^{i\vec{K}\cdot(\vec{\mathrm{i}}-\vec{\mathrm{j}})}\gamma_{\vec{K}}\ , \tag{4.5b}$$

$$\delta_{i,j}=\frac{1}{N}\sum_{\vec{K}}e^{i\vec{K}\cdot(\vec{\mathrm{i}}-\vec{\mathrm{j}})}\ , \tag{4.5c}$$

where z is the lattice coordination number and the inverse lattice sums are over the first Brillouin zone containing N points.

In this manner, Eqs. (4.4b) and (4.4a) readily lead to the following result:

$$\begin{aligned}\Gamma^{\lambda,\lambda}_{\vec{K}}(E)&=-(E+2S_\lambda zJ^{(E)})(2S_\lambda zJ^{(E)}\gamma_{\vec{K}})^{-1}\Gamma^{-\lambda,\lambda}_{\vec{K}}(E)\\&=(E+2S_\lambda zJ^{(E)})\,[E^2-(2S_\lambda zJ^{(E)})^2(1-\gamma^2_{\vec{K}})]^{-1}\ .\end{aligned} \tag{4.6}$$

Of special interest to us will be the following sums of these Fourier matrix elements:

$$\Gamma^{\lambda,\lambda}_{i,i}(E)=\frac{1}{N}\sum_{\vec{K}}\Gamma^{\lambda,\lambda}_{\vec{K}}(E)\ , \tag{4.7}$$

$$\Gamma^{-\lambda,\lambda}_{i+\delta,i}(E)=\Gamma^{-\lambda,\lambda}_{i-\delta,i}(E)=\Gamma^{-\lambda,\lambda}_{i,i+\delta}(E)=\frac{1}{N}\sum_{\vec{K}}\gamma_{\vec{K}}\,\Gamma^{-\lambda,\lambda}_{\vec{K}}(E)\ . \tag{4.8a}$$

Note that because of Eq. (3.3) we will have

$$\Gamma^{-\lambda,\lambda}_{i+\delta,i}(E)=-\Gamma^{\lambda,-\lambda}_{i+\delta,i}(E)=-\Gamma^{\lambda,-\lambda}_{i,i+\delta}(E)\ . \tag{4.8b}$$

(Here, and henceforth, the lattice position $i+\delta$ denotes one of the z nearest-neighbor positions of site i.)

From Eq. (4.6) it also readily follows that the lattices which can support interpenetrating nearest-neighbor two-sublattice antiferromagnetism cannot contain odd-order polygons whose sides are made up of nearest-neighbor vectors δ; i.e., for these lattices we have the identity

$$\frac{1}{N}\sum_{\vec{K}}(\gamma_{\vec{K}})^{2n+1}=0\ . \tag{4.9}$$

Hence from Eq. (4.6) it follows that the following matrix elements are identically vanishing:

$$\Gamma^{\lambda,\lambda}_{i,i+\delta}(E)=\frac{1}{N}\sum_{\vec{K}}\gamma_{\vec{K}}\Gamma^{\lambda,\lambda}_{\vec{K}}(E)=0, \tag{4.10a}$$

$$\frac{1}{N}\sum_{\vec{K}}\Gamma^{-\lambda,\lambda}_{\vec{K}}(E)=\Gamma^{-\lambda,\lambda}_{i,i}(E)=0\ . \tag{4.10b}$$

In conclusion, it should be emphasized that although the matrix $\Gamma(E)$ has been defined in the $(i\times\lambda)$ space, where i refers to the location of a certain lattice site and λ is the magnetic sublattice label, it is only a formal device. Physically, all we will be able to show is that, within the level of approximation inherent in the choice of a two-site Kohn neighborhood, $\Gamma^{\lambda,\lambda}_{0,0}(E)$ will be proportional to $G^{\lambda,\lambda}_{0,0}(E)$. On the other hand, within the framework of the two-site T-matrix ansatz to be employed below, it will not be possible to make a physical identification of $\Gamma^{\lambda,\lambda'}_{i,j}(E)$ for $i\neq j$.

V. TWO-SITE *T*-MATRIX ANSATZ

In complete analogy with the approximation procedure used in Paper I of the present series, we now introduce a truncated nearest-neighbor Kohn neighborhood consisting of only two sites. For simplicity we label the two sites as 0 and 1. The philosophy and the methodology of this procedure have been fully discussed in Paper I; therefore, in the following we shall only describe some of the salient details of the present calculation.

Following the prescription given in Eq. (4.2), we write the conditional thermodynamic average $G^{\lambda,\lambda}_{0,0}(E)$ by using Eq. (3.14) as follows:

$$G^{\lambda,\lambda}_{0,0}(E)=2S_\lambda\Gamma^{\lambda,\lambda}_{0,0}(E)+2S_\lambda\sum_{i,\lambda'}\sum_{j,\nu}\Gamma^{\lambda,\lambda'}_{0,i}(E)\,T^{\lambda',\nu}_{i,j}(E)\Gamma^{\nu,\lambda}_{j,0}(E), \tag{5.1}$$

where

$$T^{\lambda',\nu}_{i,j}(E)=\sum_{\{c,\sigma^\lambda_0\}}P^\lambda_0(c)\langle T^{\lambda',\nu;c}_{i,j}(E)\rangle_c\ . \tag{5.2}$$

As the truncated Kohn neighborhood of interest contains only two sites, i.e., 0 and 1, Eq. (5.1) is

$$\begin{aligned}(2S_\lambda)^{-1}G^{\lambda,\lambda}_{0,0}(E)&=\Gamma^{\lambda,\lambda}_{0,0}(E)+[\Gamma^{\lambda,\lambda}_{0,0}(E)]^2\\&\quad\times T^{\lambda,\lambda}_{0,0}(E)-[\Gamma^{-\lambda,\lambda}_{1,0}(E)]^2\,T^{-\lambda,-\lambda}_{1,1}\\&\quad-\Gamma^{\lambda,\lambda}_{0,0}(E)\Gamma^{-\lambda,\lambda}_{1,0}(E)[T^{-\lambda,\lambda}_{1,0}(E)-T^{\lambda,-\lambda}_{0,1}(E)]\ .\end{aligned} \tag{5.3}$$

Here we have made use of the fact that for the given lattice, which admits of two interpenetrating sublattices whereby in the concentrated limit all the nearest neighbors of any given site belong to the other sublattice, Eq. (4.9) holds and as such Eqs. (4.10a) and (4.10b), i.e.,

$$\Gamma^{\lambda,\lambda}_{0,1}(E)=\Gamma^{\lambda,\lambda}_{1,0}(E)=\Gamma^{-\lambda,-\lambda}_{0,1}(E)=\Gamma^{-\lambda,-\lambda}_{1,0}(E)=0 \tag{5.4}$$

and

$$\Gamma^{\lambda,-\lambda}_{0,0}(E)=\Gamma^{-\lambda,\lambda}_{0,0}(E)=\Gamma^{\lambda,-\lambda}_{1,1}(E)=\Gamma^{-\lambda,\lambda}_{1,1}(E)=0 \tag{5.5}$$

are true. Similarly, we have also made use of the identity (4.8b).

Having derived the relationship (5.3), it now remains to compute the conditional thermodynamic averages of $T_{0,0}^{\lambda,\lambda;c}(E)$, $T_{1,1}^{-\lambda,-\lambda;c}(E)$, $T_{1,0}^{-\lambda,\lambda;c}(E)$, and $T_{0,1}^{\lambda,-\lambda;c}(E)$. In the following we shall first derive expressions for these matrix elements in terms of the appropriate matrix elements of V^c and Γ and next we shall carry out the conditional averaging.

Because our interest here is in considering only those configurations for which the site 0 is known to be occupied by a magnetic atom belonging to the sublattice λ, we put $\sigma_0^\lambda = 1$. However, for site 1 we still have to explicitly use the magnetic occupation operator $\sigma_1^{\lambda'}$, which will be unity when the site 1 happens to have a magnetic atom belonging to sublattice λ' on it. Otherwise, $\sigma_1^{\lambda'}$ will be zero. Within this framework, and using the notation of Eq. (2.5) and the relationship (3.3), we can write down the relevant matrix elements of $V^c(E)$ from their defining equation (3.6b), i.e.,

$$V_{0,0}^{\lambda,\lambda;c}(E) = 2S_\lambda(J\sigma_1^{-\lambda} - J^{(E)}) = -V_{1,1}^{-\lambda,-\lambda;c}(E) , \qquad (5.6)$$

$$V_{0,1}^{\lambda,-\lambda;c}(E) = -V_{1,0}^{-\lambda,\lambda;c}(E) = V_{0,0}^{\lambda,\lambda;c}(E) . \qquad (5.7)$$

Taking matrix elements of both sides of the matrix equation (3.17) and using the foregoing we get

$$\begin{aligned} T_{0,0}^{\lambda,\lambda;c}(E) &= -T_{1,0}^{-\lambda,\lambda;c}(E) = T_{0,1}^{\lambda,-\lambda;c}(E) = -T_{1,1}^{-\lambda,-\lambda;c}(E) \\ &= V_{0,0}^{\lambda,\lambda;c}(E)\{1 - V_{0,0}^{\lambda,\lambda;c}(E) \\ &\quad \times [\Gamma_{0,0}^{\lambda,\lambda}(E) - \Gamma_{0,0}^{-\lambda,-\lambda}(E) + 2\,\Gamma_{1,0}^{-\lambda,\lambda}(E)]\}^{-1} . \qquad (5.8) \end{aligned}$$

This result is interesting in that it belatedly justifies our choice of a single arbitrary parameter $J^{(E)}$ for arranging the equality of the Green's functions $(2S_\lambda)^{-1} G_{0,0}^{\lambda,\lambda}(E)$ and $\Gamma_{0,0}^{\lambda,\lambda}(E)$. Clearly, when the conditional average of any one of the four matrix elements, e.g., $T_{0,0}^{\lambda,\lambda}(E)$, given in Eq. (5.8), is made vanishing, according to Eq. (5.3) it follows that

$$G_{0,0}^{\lambda,\lambda}(E) = 2S_\lambda\, \Gamma_{0,0}^{\lambda,\lambda}(E) . \qquad (5.9)$$

The central result of the present section, therefore, is the expression for $J^{(E)}$ which is obtained from the ansatz:

$$T_{0,0}^{\lambda,\lambda}(E) = 0 , \qquad (5.10)$$

where $T_{0,0}^{\lambda,\lambda}(E)$ is as given by Eq. (5.8) with the conditional averaging prescribed in Eq. (5.2).

VI. FREQUENCY MOMENTS OF DENSITY OF STATES

Having derived the ansatz (5.10) for the computation of $J^{(E)}$, we are now in a position to examine the frequency moments of the density of states, $\rho_\lambda(\omega)$, where

$$2S_\lambda \rho_\lambda(\omega) = \frac{1}{2\pi}\int_{-\infty}^{+\infty} \langle [S_0^+(\lambda, t), S_0^-(\lambda, t')]_-\rangle_{(0,\lambda)} \times e^{i\omega(t-t')}\, d(t-t') . \qquad (6.1)$$

From the definition of the Green's function $G_{0,0}^{\lambda,\lambda}(\omega)$ [see Eqs. (3.1) and (4.2)] it follows that

$$G_{0,0}^{\lambda,\lambda}(E) = 2S_\lambda \int_{-\infty}^{+\infty} \frac{\rho_\lambda(\omega)}{E-\omega}\, d\omega, \quad \mathrm{Im}E > 0 . \qquad (6.2)$$

Because the frequency moments of the density of states $\rho_\lambda(\omega)$ are proportional to equal time thermodynamic averages, which are finite, for ReE $\gg \omega$ we can expand the right-hand side of Eq. (6.2) in inverse powers of E to get

$$E G_{0,0}^{\lambda,\lambda}(E) = 2S_\lambda \sum_{n=0}^{\infty} M_n^\lambda / E^n = 2S_\lambda E \Gamma_{0,0}^{\lambda,\lambda}(E) , \qquad (6.3)$$

where M_n^λ is the nth-frequency moment of $\rho_\lambda(\omega)$, i.e.,

$$M_n^\lambda = \int_{-\infty}^{+\infty} \rho_\lambda(\omega)\, \omega^n\, d\omega . \qquad (6.4)$$

Let us next write out Eq. (5.10) in detail so as to calculate $J^{(E)}$, which in turn will determine $\Gamma_{0,0}^{\lambda,\lambda}(E)$. Using Eqs. (5.6) and (5.8), Eq. (5.10) gives

$$\begin{aligned} &m\, 2S_\lambda(J - J^{(E)})\{1 - 2S_\lambda(J - J^{(E)})[\Gamma_{0,0}^{\lambda,\lambda}(E) - \Gamma_{0,0}^{-\lambda,-\lambda}(E) + 2\,\Gamma_{1,0}^{-\lambda,\lambda}(E)]\}^{-1} \\ &\qquad + (1-m)\, 2S_\lambda(-J^{(E)})\{1 - 2S_\lambda(-J^{(E)})[\Gamma_{0,0}^{\lambda,\lambda}(E) - \Gamma_{0,0}^{-\lambda,-\lambda}(E) + 2\,\Gamma_{1,0}^{-\lambda,\lambda}(E)]\}^{-1} = 0 . \qquad (6.5) \end{aligned}$$

The first term on the left-hand side of Eq. (6.5) is contributed by the magnetic occupancy of the site 1. Note that because site 1 is a nearest neighbor of site 0, which is known to have a magnetic atom belonging to sublattice λ, the probability of finding a magnetic atom belonging to sublattice $-\lambda$ at site 1 is equal to m [see Eq. (2.9b)]. The occupancy of site 1 by an atom which is either nonmagnetic or is of variety $+\lambda$ leads to the second term on the left-hand side of Eq. (6.5).

It is convenient to recast Eq. (6.5) into the following compact form:

$$\eta(E) = J^{(E)}/J = [mz - 2F(E)][z - 2F(E)]^{-1} , \qquad (6.6)$$

$$F(E) = 1 - \frac{E^2}{N}\sum_{\vec{K}} \{E^2 - [2S_\lambda zJ\eta(E)]^2(1 - \gamma_{\vec{K}}^2)\}^{-1} . \qquad (6.7)$$

Expanding $\eta(E)$, i.e.,

$$\eta(E)=\sum_{n=0}^{\infty}(2S_\lambda Jz)^n\eta^{(n)}/E^n\ , \tag{6.8}$$

and self-consistently determining $\eta^{(n)}$ from Eqs. (6.6)–(6.8) by comparing coefficients of various powers of E^{-1}, and then using the result (6.3), we find the following results for the first five moments:

$$\Delta(0)=1\ , \tag{6.9a}$$

$$\Delta(1)=m\ , \tag{6.9b}$$

$$\Delta(2)=m^2-m/z\ , \tag{6.9c}$$

$$\Delta(3)=m^3+(-3m^3+2m^2)/z+(2m^3-2m^2)/z^2\ , \tag{6.9d}$$

$$\Delta(4)=m^4+(-6m^4+4m^3)/z+[m^4\phi(4)+8m^4-8m^3]/z^2+(-4m^4+4m^3)/z^3\ , \tag{6.9e}$$

where

$$M_n^\lambda=(2S_\lambda zJ)^n\Delta(n)\ , \tag{6.10a}$$

$$\begin{aligned}\phi(4)=z^2\frac{1}{N}\sum_{\vec{K}}\gamma_{\vec{K}}^4&=2.5, &&\text{sc;}\\ &=3.375, &&\text{bcc;}\\ &=3.75, &&\text{fcc .}\end{aligned} \tag{6.10b}$$

It is interesting to compare these results with those obtained for the randomly dilute ferromagnet. While the first two moments of the density of states, i.e., M_0^λ and M_1^λ, coincide for these two systems, higher-order moments are different in the next to the leading order in z^{-1}. The identity of the leading-order terms in z^{-1} can, of course, be anticipated from the knowledge that this term is given exactly by a correct mean-field-theory approximation, and within such an approximation both the dynamics and the statics of the two systems are identical (e.g., for zero external field, the magnitudes of the sublattice magnetization and the Néel temperature are the same as ferromagnetic magnetization and the Curie temperature, respectively).

To compare the results (6.9a)–(6.9e) obtained from the use of the present approximate theory with the corresponding "exact" results, it is necessary first to make precise the meaning of the term exact. Because the use of the RPA-like decoupling, implicit in going from Eq. (3.2) to Eq. (3.4), is equivalent to introducing a certain approximation for the dynamics and thermostatics of the system even in the concentrated limit $m=1$, Eqs. (6.9a)–(6.9e) portray the complicated interplay of the approximations introduced in the study of two distinct complexities of the problem; namely, the many-body nature of the interactions and the complications caused by randomness. Because our interest in the present work is in the study of the latter aspect of the problem only, we first give an explicit representation of a model system for which our decoupled Green's functions [i.e., as given in Eq. (3.4)] can be considered to be exact.

The simplest such model is the zero-temperature noninteracting spin-wave-theory model whereby

$$\begin{aligned}&S_g^+(\lambda,t)\to(2S_\lambda)^{1/2}a_g(t), \quad S_g^-(\lambda,t)\to(2S_\lambda)^{1/2}a_g^\dagger(t);\\ &S_g^-(-\lambda,t)\to(2S_\lambda)^{1/2}b_g(t), \quad S_g^+(-\lambda,t)\to(2S_\lambda)^{1/2}b_g^\dagger(t);\\ &S_g^z(\lambda,t)\to S_\lambda-a_g^\dagger(t)a_g(t), \quad S_g^z(-\lambda,t)\to -S_\lambda+b_g^\dagger(t)b_g(t).\end{aligned} \tag{6.11}$$

Here the operators a and b obey the Bose commutation relations

$$[c_g,c_f]_-=[c_g^\dagger,c_f^\dagger]_-=0,\quad [c_g,c_f^\dagger]_-=\delta_{g,f}\qquad \text{for } c=a,b \tag{6.12}$$

and

$$[a_g,b_f]_-=[a_g^\dagger,b_f^\dagger]_-=[a_g,b_f^\dagger]_-=0\ . \tag{6.13}$$

In this representation the Hamiltonian $\mathcal{H}^c$ is to be taken

$$\mathcal{H}^c=\text{const}+2S_\lambda\sum_{f_1,f_2}{}'\sigma_{f_1}^{\lambda}\sigma_{f_2}^{-\lambda}\times(a_{f_1}^\dagger a_{f_2}+b_{f_1}^\dagger b_{f_1}+a_{f_1}^\dagger b_{f_2}^\dagger+a_{f_1}b_{f_2})\ . \tag{6.14}$$

In terms of the analysis presented in the preceding sections, the Green's function $\langle\langle a_0(t); a_0^\dagger(t')\rangle\rangle$ is readily seen to be the relevant approximation to the conditionally averaged thermodynamic Green's function $G_{0,0}^{\lambda;\lambda}(t-t')$. Because for this model the counterpart of the density of states, $\rho_\lambda(\omega)$, is

$$\rho_\lambda(\omega)=\frac{1}{2\pi}\int_{-\infty}^{+\infty}\langle[a_0(t),a_0^\dagger(t')]_-\rangle_{(0,\lambda)}\times e^{i\omega(t-t')}d(t-t')\ , \tag{6.15}$$

the exact moments of the density of states may be evaluated by using the well-known formal prescription

$$M_n^\lambda=\langle[[\cdots[a_0,\mathcal{H}^c]_-,\cdots,\mathcal{H}^c]_-,[\cdots[a_0^\dagger,\mathcal{H}^c]_-,\cdots,\mathcal{H}^c]_-]_-\rangle_{(0,\lambda)}\ , \tag{6.16}$$

where the commutators of a_0 and $a_0^\dagger$ with $\mathcal{H}^c$ are repeated i times and $n-i$ times, respectively. Here $0\leqslant i\leqslant n$. The computations are straightforward though tedious and we readily ascertain that the results (given by our approximate theory) for the first four moments are exact. The exact fifth moment, however, is different from the approximate one given in Eq. (6.9e), i.e.,

$$[\Delta(4)]_{\text{exact}} = m^4 + (-6m^4 + 4m^3)/z + [11m^4 - 12m^3 + m^3\phi(4) + m^2]/z^2 + (-6m^4 + 7m^3 - m^2)/z^3 . \quad (6.17)$$

A term-by-term comparison of the right-hand side of this equation with that of Eq. (6.9e) reveals that the approximate and the exact results agree in the two leading terms in the z^{-1} expansion. This situation is analogous to that encountered for the dilute ferromagnet. Stretching this analogy further we might expect that perhaps the present ansatz would reproduce all the moments, i.e., M_n^λ for arbitrary n, exactly to the leading two orders in z^{-1}. It is to the investigation of this question that we now address ourselves.

Let us first inquire as to the formal structure of the exact expression for the nth frequency moment. To determine this we first need to find $\sigma_g^\lambda D^n a_g$, where

$$D \equiv i\,\frac{d}{dt} , \quad (6.18a)$$

that is,

$$\sigma_g^\lambda D a_g = \sigma_g^\lambda [a_g, \mathcal{H}^c]_- = \sigma_g^\lambda (2S_\lambda) \sum_{f_1} J^{\lambda,-\lambda}(gf_1)\, \sigma_{f_1}^{-\lambda}(a_g + b_{f_1}^\dagger) ,$$

$$\sigma_g^\lambda D^2 a_g = \sigma_g^\lambda (2S_\lambda)^2 \sum_{f_1}\sum_{f_2} J^{\lambda,-\lambda}(gf_1) \times [J^{\lambda,-\lambda}(gf_2)\, \sigma_{f_1}^{-\lambda}\sigma_{f_2}^{-\lambda}(a_g + b_{f_2}^\dagger) - J^{\lambda,-\lambda}(f_2 f_1)\, \sigma_{f_1}^{-\lambda}\sigma_{f_2}^{\lambda}(a_{f_2} + b_{f_1}^\dagger)] , \quad (6.18b)$$

etc. Next we need the commutator $\sigma_g^\lambda [D^n a_g, a_g^\dagger]_-$, which contains 2^{n-1} terms. (Note that because a and b operators commute, the total number of terms for this problem is only half what it was for the ferromagnetic case.) Out of these 2^{n-1} terms there is a unique term $B(n)$, which involves spatial correlations of n spatial locations, i.e.,

$$B(n) = (2S_\lambda)^n \sum_{f_1} \cdots \sum_{f_n} J^{\lambda,-\lambda}(gf_1) \cdots J^{\lambda,-\lambda}(gf_n) \times \langle \sigma_{f_1}^{-\lambda} \cdots \sigma_{f_n}^{-\lambda} \rangle_{(g,\lambda)} . \quad (6.19)$$

Because of the presence of $J^{\lambda,-\lambda}(gf_i)$ factors, none of the n spatial locations $f_1, \ldots, f_n$ can coincide with g. Therefore, with the assumption of complete spatial randomness we readily find that

$$B(n) = (2S_\lambda Jz)^n m^{n-1}\left[m\left(1 - \frac{n(n-1)}{2z}\right) + \frac{n(n-1)}{2z} + O\left(\frac{1}{z^2}\right)\right] . \quad (6.20)$$

Note that this result is exactly analogous to the corresponding result obtained for the dilute ferromagnet [see Eq. I(A6)].

Unlike the term $B(n)$, the structure of the remaining $(2^{n-1} - 1)$ terms for the present case differs from that obtained for the ferromagnet. The major difference lies in the fact that here the remaining terms can give contributions with positive as well as negative signs, which are of the order $(2S_\lambda Jz)^n/z$. [Recall that in the ferromagnetic case, to this order, the remaining terms in M_n^λ contributed only with a positive sign. Because the total number of such terms was equal to the number of times we can arrange two distinct sites, out of a total of n, to be identical, i.e., $\frac{1}{2}n(n-1)$, the relevant summation was trivial.] Here, for the case of even n, i.e., $n = 2\nu$, of the $\nu(2\nu - 1)$ terms only ν^2 are negative. The remaining $\nu^2 - \nu$ are positive. Consequently, the total contribution of the remaining terms in the leading order in z^{-1} is negative, i.e.,

$$C(n = 2\nu) = (2SJz)^n \left[\frac{m^{\nu-1}}{z}(-\nu) + O\left(\frac{1}{z^2}\right)\right] . \quad (6.21a)$$

If, on the other hand, n is odd, i.e., $n = 2\nu + 1$, then of the $\nu(2\nu + 1)$ terms ν^2 are positive and $\nu^2 + \nu$ are negative. The net result of this is that the leading contribution to the moment M_n^λ for the case $n = 2\nu + 1$ is again negative and it formally resembles the right-hand side of Eq. (6.21a), i.e.,

$$C(n = 2\nu + 1) = (2SJz)^n \left[\frac{m^{\nu-1}}{z}(-\nu) + O\left(\frac{1}{z^2}\right)\right] . \quad (6.21b)$$

Combining the results (6.20), (6.21a), and (6.21b) we have

$$M_n^\lambda = B(n) + C(n) + \text{remainder} , \quad (6.22)$$

which gives

$$M_{2\nu}^\lambda = (2SJz)^{2\nu} m^{2\nu-1}\{m[1 - \nu(2\nu - 1)/z] + 2\nu(\nu - 1)/z + O(1/z^2)\} , \quad (6.23a)$$

$$M_{2\nu+1}^\lambda = (2SJz)^{2\nu+1} m^{2\nu}\{m[1 - \nu(2\nu + 1)/z] + 2\nu^2/z + O(1/z^2)\} . \quad (6.23b)$$

The computation of the corresponding results from the approximate ansatz (6.5) is also straightforward. Using Eqs. (6.6)–(6.8) we note that in an order-by-order series expansion in powers of z^{-1} we can write

$$\eta^{(n)} = \sum_{p=0}^{\infty} \eta_p^{(n)} z^{-p} , \quad (6.24)$$

and in this way self-consistently calculate $\eta_p^{(n)}$. The relevant results are

$$\eta^{(0)}=m\,,\quad \eta^{(2\nu+1)}=0\;; \tag{6.25a}$$

$$\eta_0^{(2\nu)}=0,\quad \eta_1^{(2\nu)}=2m^{2\nu}(1-m)\;. \tag{6.25b}$$

Note that Eq. (6.25a) gives $\eta^{(0)}$ and $\eta^{(2\nu+1)}$ to all orders in z^{-1}. On the other hand, Eq. (6.25b) records the result for $\eta^{(2n)}$ only to the two leading orders in z^{-1}. This is, however, quite sufficient for the present purposes because higher-order results contribute to the $O(1/z)^2$ terms on the right-hand side of Eqs. (6.22a) and (6.22b). With this knowledge we readily derive the given results in Eqs. (6.23a) and (6.23b) by using the power expansion (6.3), with the additional embellishment that each term has its own z^{-1} expansion series also.

VII. SUBSTITUTIONAL SHORT-RANGE ORDER

The foregoing analysis assumed complete substitutional randomness of the magnetic and the nonmagnetic atoms. In practice, however, the alloy will contain some residual SRO with the result that if we are given a magnetic atom on the site 0, the likelihood of finding another (magnetic atom) on the neighboring site 1 will not be strictly equal to the relative magnetic concentration m. Rather, if the magnetic-nonmagnetic SRO is α_0 then this probability mQ^{-1} will be

$$mQ^{-1}=m+(1-m)\,\alpha_0\;. \tag{7.1}$$

Of course, given that the site 0 has a magnetic atom, the probability that the neighboring site 1 has a nonmagnetic atom is Q' such that

$$Q'=(1-m)(1-\alpha_0)\;. \tag{7.2}$$

Clearly mQ^{-1} and Q' should add up to unity irrespective of the amount of SRO (α_0), and they do.

In terms of the occupation operators, σ_i^λ, etc., the significance of the SRO is as follows: Define a magnetic occupation operator $\hat{m}_i$ by the relation

$$\hat{m}_i=\sigma_i^\lambda+\sigma_i^{-\lambda}\;. \tag{7.3}$$

Clearly, the eigenvalue of the magnetic occupation operator is $+1$ if the relevant site i is occupied by a magnetic atom (note that whether such an atom is a member of the up or the down antiferromagnetic sublattice is not material to this discussion). However, if the site i has a nonmagnetic atom, then $\hat{m}_i=0$ but the nonmagnetic occupation operator $\hat{n}_i$ must be unity for this case. Therefore, we always have

$$\hat{m}_i+\hat{n}_i=1\;. \tag{7.4}$$

The SRO α_0 is now readily defined from the relation

$$\langle\hat{m}_i\hat{n}_{i+\delta}\rangle=m(1-m)(1-\alpha_0)\;. \tag{7.5a}$$

The substitutional correlation on the left-hand side is a nonrestricted thermodynamic correlation between the nonmagnetic and the magnetic occupation of an arbitrary pair of neighboring sites. The partial conditional correlation (which assumes *a priori* that the site i is occupied by a magnetic atom), on the other hand, is related to the SRO as follows:

$$\langle\hat{m}_i\hat{n}_{i+\delta}\rangle_{(i,m)}=Q'=(1-m)(1-\alpha_0)\quad\text{(site } i \text{ magnetic)}\;. \tag{7.6a}$$

The remaining two correlation functions are now readily given, i.e.,

$$\langle\hat{m}_i\hat{m}_{i+\delta}\rangle=m^2+m(1-m)\,\alpha_0\;, \tag{7.5b}$$

$$\langle\hat{m}_i\hat{m}_{i+\delta}\rangle=m+(1-m)\,\alpha_0=Q^{-1}m\quad\text{(site } i \text{ magnetic)}\;. \tag{7.6b}$$

The introduction of the SRO clearly does not affect the treatment of Secs. I–IV. Only the details of the averaging process over site 1 (see Secs. V and VI) are affected. As such, Eq. (6.5) gets modified to read as follows:

$$\begin{aligned}&mQ^{-1}\,2S_\lambda(J-J^{(E)})\{1-2S_\lambda(J-J^{(E)})[\Gamma_{0,0}^{\lambda,\lambda}(E)-\Gamma_{0,0}^{-\lambda,-\lambda}(E)+2\,\Gamma_{1,0}^{-\lambda,\lambda}(E)]\}^{-1}\\&\qquad=Q'\,2S_\lambda J^{(E)}\{1+2S_\lambda J^{(E)}[\Gamma_{0,0}^{\lambda,\lambda}(E)-\Gamma_{0,0}^{-\lambda,-\lambda}(E)+2\,\Gamma_{1,0}^{-\lambda,\lambda}(E)]\}^{-1}\;.\end{aligned} \tag{7.7}$$

The net effect of this modification is to change the actual magnetic concentration m to an effective magnetic concentration equal to mQ^{-1}. When this transformation is introduced into Eq. (6.5), it becomes identical to Eq. (7.7).

In view of the above, the results of Sec. VI can in large part be taken over for the case with nonzero SRO. For example, now the system Néel temperature will scale with the magnetic concentration and the SRO as follows:

$$T_c(m)/T_c(1)=(zmQ^{-1}-2)/(z-2)\;. \tag{7.8}$$

This means that even if the actual concentration m is less than the critical minimum value m_0 we can still get a correlated ground state with antiferromagnetic LRO as long as

$$mQ^{-1}>2/z\;. \tag{7.9}$$

But this can always be achieved by building a suitable clustering correlation $\alpha_0 > 0$ among the magnetic atoms. For example, if such magnetic clustering is perfect, i.e., $\alpha_0 = +1$, then $Q = m$ and therefore in three dimensions Eq. (7.9) is always satisfied. Heuristically, this is readily understandable, for if α_0 is $+1$, all the magnetic atoms will cluster together. This way, as long as $m \neq 0$ (if m were equal to zero the discussion of the system magnetism in the present macroscopic sense would be meaningless), all the (macroscopic number of) magnetic atoms would sit in one region of the lattice and that region would be filled perfectly with only the magnetic atoms. Clearly, then, such a segregated alloy would be macroscopically ordered and its properties would approach that of a perfect magnetic system (of its size).

On the other hand, if $\alpha_0 < 0$, then the system would attempt to keep magnetic sites apart from each other (also, of course, it would treat the nonmagnetic sites similarly) and if perchance $m = \frac{1}{2}$, that is, if the system consisted of half magnetic and half nonmagnetic atoms, then because of the choice of the lattice (which admits two nearest-neighbor interpenetrating sublattices) the system would be able to keep the magnetic and nonmagnetic sites apart perfectly when $\alpha_0 = -1$. In other words, it would then order in an AB pattern such that a magnetic atom will see only nonmagnetic atoms (as its nearest neighbors) and vice versa. Such a system would then be completely nonmagnetic. This heuristic argument is supported by the present result, for then [see Eqs. (7.1) and (7.8)] we find that $T_c(m)$ becomes negative. The negative Néel temperature is a meaningless concept. It only implies here that in such a situation the antiferromagnetic LRO does not obtain at all. For $m \neq \frac{1}{2}$, the occurrence of a completely perfect alternating AB type of order becomes a physical impossibility. Other more complex stoichiometric situations can, of course, occur but we will not discuss those here. It is, however, clear that by suitable manipulation of the SRO, if the magnetic concentration is less than $(\frac{1}{2} + 1/z)$, the system can be kept from attaining a state with antiferromagnetic LRO at all temperatures, for then the inequality

$$m + (1-m)\,\alpha_0 < 2/z \tag{7.10}$$

can be satisfied. However, if $m > (\frac{1}{2} + 1/z)$, the system will always have a finite Néel temperature, for then there exists no effective way of hiding the magnetic atoms from each other such that on the average each atom has two, or less, of its nearest-neighbor atoms magnetic.

It is interesting to note that the smallest value of α_0 for which the system has no nonzero critical concentration is equal to $+(2/z)$. For this value of the SRO parameter we have

$$T_c(m)/T_c(1) = m(1 - 2/z), \qquad \alpha_0 = 2/z\ . \tag{7.11}$$

To conclude this section, we should like to prove that if the substitutional correlations among the two varieties of atoms are of a particularly simple variety such that the conditional correlation referring to p distinct spatial locations $1, 2, \ldots p$, i.e.,

$$\langle \hat{m}_1 \hat{m}_2 \cdots \hat{m}_p \rangle_{(0,\lambda)} = \hat{F}(1, 2, \ldots, p)\ , \tag{7.12}$$

decouples into the two-point nearest-neighbor conditional correlations in the manner prescribed by the cumulant decoupling,[9] then the accuracy of the results in the presence of the nearest-neighbor SRO is only slightly inferior to the case when the SRO is vanishing.

Let us first make the above statement more precise. First, let us explain the conditional aspect of the correlation given in Eq. (7.12). Here, for convenience, we assume that the site 0 is known to be occupied by a magnetic atom (of, say, λ-sublattice variety). Second, because of the fact that if any of the sites $1-p$ are the same, then the operator identity

$$(\hat{m}_i)^2 = \hat{m}_i \tag{7.13a}$$

causes the correlation (7.12) to reduce to that of one lower order, we only need to consider the case where all the given locations $1-p$ are distinct. Of course, here $p \leq z$. Now, according to the treatment presented in Sec. VI, correlations of the type (7.12) contribute in the leading order in the z^{-1} expansion. Moreover, in the lowest-order cumulant decoupling approximation, we have

$$\hat{F}(1, 2, \ldots, p) \cong \langle \hat{m}_1 \rangle \langle \hat{m}_2 \rangle \cdots \langle \hat{m}_p \rangle\ . \tag{7.13b}$$

Because each of the conditional averages on the right-hand side of Eq. (7.13b) refers to the neighboring sites of the site 0, Eq. (7.13b) reduces to the following:

$$\hat{F}(1, 2, \ldots, p) = (mQ^{-1})^p, \qquad p \leq z\ . \tag{7.14}$$

Recall that mQ^{-1} is the effective magnetic concentration in the presence of the SRO and that according to the present ansatz this replacement completely specifies the extension of the theory to the nearest-neighbor SRO case.

In light of the foregoing, it can be assumed as having been established that for sufficiently idealized structure of the substitutional correlations (namely, a Gaussian-like structure whereby the lowest-order cumulant decoupling obtains), the present theory gives all the frequency moments exactly to the leading order in z^{-1}. As regards the situation for the first few moments, it is straightforward (though tedious) to corroborate that the first three moments of the single-site density of states are now preserved exactly.

The significance of the above results is the following: The present theory is exact in the limit that the spatial locations are perfectly correlated, and as a result we get a system which can be described by two interpenetrating nearest-neighbor sublattices. Moreover, the accuracy of the results is fairly high in the limit of vanishing substitutional correlations, especially if the coordination number of the lattice, z, is large. In between these limits, a reasonable interpolation to the results for arbitrary SRO is provided by the present theory as long as the many-atom clustering effects can be approximated by neighboring atom (two-atom) clusters.

VIII. NONINFINITE z

The accuracy of the theory presented in this paper has been justified to the two leading orders in an inverse z expansion. Because the molecular-field and virtual-crystal types of mean-field approximations are thought to be exact only to the leading order in z^{-1} (note that our mass operator agrees with that given by a proper combination of these theories to the leading order in z^{-1}), the present theory can be thought to give the first-order correction to the mean-field approximation in the asymptotic z^{-1} regime. The question that arises is how good the results of the present theory can be expected to be for systems where the effective magnetic-lattice coordination number z is of the order of 10.

Clearly, 10 is not a large number in the statistical sense. Therefore for $z \sim 10$, the statistical fluctuation in the number of magnetic nearest neighbors of a given magnetic atom can be expected to be large and hence the magnetic concentration of $2/z + \epsilon$, where $\epsilon \to +0$, cannot guarantee the presence of at least two magnetic nearest neighbors for all magnetic atoms. This means that even when m is $2/z + \epsilon$, the existence of a macroscopic network of nearest-neighbor exchange-coupled links cannot be assumed. Because the antiferromagnetic LRO cannot occur in the absence of such a network, the critical concentration m_0 must be larger than $2/z$.

Unfortunately, an estimate of the amount by which m_0 exceeds $2/z$ cannot be given. In the asymptotic large-z limit, corrections to the result $m_0 \sim 2/z$ which are in the nature of a power series beginning with the order z^{-2} can, however, be expected. Precisely how rapidly such a series converges, or indeed whether it converges at all, we are not able to establish.

To conclude, it should be mentioned that a theory which, unlike the present one, does not make the quasiclassical assumption about the antiferromagnetic ground state would in all likelihood give additional spin-dependent corrections to m_0. These corrections can, however, be expected to decrease with the increase in the magnitude of the spin S.

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