J. Phys. C: Solid State Phys., 17 (1984) 5367–5384. Printed in Great Britain

The coherent potential approximation and the diagrammatic 1/Z expansion for an anisotropic spin-1 paramagnet

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Received 18 October 1983, in final form 20 March 1984

Abstract. The magnetic Green functions in the paramagnetic phase of a dilute or a binary alloy system are calculated for arbitrary concentrations of magnetic ions. The calculation employs a diagrammatic perturbation formalism based on the use of semi-invariants. Specifically, the theory is applied to $S = 1$ systems in both the easy-axis and the easy-planar cases. The approximation made in the theory is basically to consider the ions as separate entities embedded in an effective medium, which is established in a self-consistent fashion. The calculated self-energy of the Green functions contains both the CPA contributions, due to the inhomogeneities, and the contributions from the fourth-order semi-invariants of the single spins appearing in order $1/Z$ in the high-density expansion. The behaviour of the effective single–site Green function is analysed in detail. The spurious divergence of this quantity in the order $(1/Z)^0$, which leads to unphysical results in the dilute limit, is found to be much suppressed.

1. Introduction

The coherent-potential approximation (CPA) for a disordered crystalline system has been applied successfully to a wide range of problems (for a review see Elliott *et al* 1974). Here, we shall consider spin systems in the paramagnetic phase. The coupling between the spins is an isotropic Heisenberg interaction, and the single-spin states are split by the crystalline field, which is assumed to be independent of environmental spin disorder. In the homogeneous case these systems support boson-like excitations which are reasonably well defined at low temperatures. A CPA approach which has been utilised in previous works is the cumulant expansion of the averaged Green functions, developed by Yonezawa (1968). The method has been applied to dilute spin systems by Schmidt (1974) and by Lage and Stinchcombe (1977), whose theory has been extended to the case of a binary alloy system by Whitelaw (1981). In their theory Lage and Stinchcombe included a single-site term depending on the environmental disorder. If this inhomogeneous term is neglected their CPA result reduces to that obtained by Schmidt (1974), and the Green function of the non-dilute system becomes the one obtained in the random-phase approximation (RPA). With the use of the RPA Green function for the paramagnetic spin system the effect of dilution is the same, within CPA, as obtained for a phonon system when introducing diagonal disorder with $\varepsilon \rightarrow -\infty$ (ε is the mass defect

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factor: see for instance Elliott *et al* 1974). The CPA result is well behaved in the case of phonons but for a paramagnetic spin system the combined RPA–CPA approach leads to an unphysical behaviour in the dilute limit. For instance if the ground state of the single spins is degenerate, the static susceptibility is predicted to diverge at a temperature which stays finite in the limit $c \rightarrow 0$: see also the discussion by Wang (1973). This spurious divergence indicates the need for a more accurate treatment of the spin system than is offered by the RPA; in contrast to a system of coupled harmonic oscillators, each of the single spins can only be excited into a few different states. Fluctuations between these states have direct consequences for the Green functions which are neglected in RPA.

Lage and Stinchcombe (1977) succeeded in removing the unacceptable behaviour of the theory in the dilute limit by the introduction of a conditional inhomogeneous term in the Hamiltonian, which in the comparison with a phonon system corresponds to the introduction of off-diagonal disorder. The magnitude of the inhomogeneous term was determined such that the Green function in the concentrated limit of $c = 1$ coincided with the result of the correlated-effective-field theory of Lines (1974, 1975). The inhomogeneous single-site term in the non-dilute case was introduced by Lines in a phenomenological way, in order to account for the effects of spin fluctuations on the Green function. In this theory the thermal spin fluctuations give rise to a renormalisation of the coupling parameters, but they do not contribute directly to the lifetime of the excitations. However, in paramagnetic spin systems the lifetime effects of thermal and static disorder might easily be comparable (see for instance Jensen 1979). Besides avoiding the use of a phenomenological procedure we shall here include thermal lifetime effects by considering instead the leading-order perturbative correction to the RPA Green function.

The perturbative procedure we use is a linked-cluster expansion of the time-ordered Green functions. In this the unperturbed averages of spin operators occur grouped into cumulant averages or semi-invariants in a systematic expansion of the Green functions in powers of $1/Z$, the inverse coordination number. The method was applied to an isotropic ferromagnet by Vaks *et al* (1968), and has now been applied to a number of systems; by Stinchcombe (1973) to the $S = \frac{1}{2}$ Ising model, by Yang and Wang (1975) and Psaltakis and Cottam (1982) to the $S = 1$ easy-axis magnet, while Bak (1975) has considered lifetime effects in the $S = 1$ easy-planar paramagnet. In principle, the semi-invariants are straightforwardly calculated using the Wick-like theorem established by Care and Tucker (1977), but in practice the method becomes quite cumbersome if the number of single-spin states or spin-operators is not small. The theory we shall present is valid for an arbitrary value of S, but the actual calculation of the semi-invariants is carried through only for $S = 1$.

The zero-order approximation in $1/Z$ is identical with the RPA. In order $1/Z$ the four-spin-operator semi-invariants are introduced, and they contribute to the real and imaginary parts of the self-energy. In this order the spins are treated as separate entities placed in an effective medium. This way of characterising the approximation shows it to be equivalent to the CPA in the inhomogeneous case. In order to combine the $1/Z$ Green function with the CPA we utilise directly this equivalence. Instead of following the procedure developed by Yonezawa (1968) we arrange the diagrams in the cumulant expansion in a way which allows us to consider separately the single sites interacting with the configurationally averaged effective medium. Because of this arrangement it turns out to be a straightforward matter to include the $1/Z$ contributions in the CPA Green functions, both in the dilute case and in the case of an arbitrary binary alloy system.

In § 2 we develop the CPA theory in the general case, and the results are expressed in terms of the single-site Green function. In § 3 we calculate this Green function when $S = 1$, and compare the static behaviour predicted by the various approximation schemes. In the last section we discuss the results of our calculation.

2. General theory

We shall be concerned with systems in which two kinds of atoms are distributed on a lattice in a random fashion. To start with we shall assume that only the one kind of atom with concentration c has a non-zero spin S. The single-spin properties are given by the Hamiltonian $\mathcal{H}_0$, which is assumed to be independent of the various environments occurring in the lattice. The spins are coupled by the perturbative Hamiltonian:

$$\mathcal{H}_1 = -\tfrac{1}{2}\sum_{i\neq j} c_i c_j J(ij)\,\boldsymbol{S}_i \cdot \boldsymbol{S}_j \tag{1}$$

where $c_i = 1$ if the ith site is occupied by a magnetic atom, otherwise $c_i = 0$. In order to simplify the notation we shall consider only one spin components and we suppress the corresponding indices in the following.

The two-site Green functions are defined as the τ-ordered ensemble averages

$$G(ij, \tau_1 - \tau_2) = \langle T_\tau c_i S_i(\tau_1) c_j S_j(\tau_2)\rangle \tag{2}$$

where $S_i(\tau)$ is the spin operator of the ith site in the interaction representation, and $-\beta < \tau_1 - \tau_2 < \beta$ with $\beta = 1/k_B T$. The Fourier transforms are defined in terms of the Matsubara frequencies $\omega_n = 2\pi n/\beta$ (n is an integer)

$$\langle G(\boldsymbol{q}, \mathrm{i}\omega_n)\rangle = \frac{1}{N}\sum_{i,j} \exp(\mathrm{i}\boldsymbol{q}\cdot\boldsymbol{R}_{ij}) \int_0^\beta G(ij, \tau)\exp(\mathrm{i}\omega_n\tau)\,\mathrm{d}\tau. \tag{3}$$

This definition includes the configurational averaging (the sum over i) as indicated by $\langle\ \rangle$, and $\langle G(\boldsymbol{q}, \omega)\rangle$ is obtained from $\langle G(\boldsymbol{q}, i\omega_n)\rangle$ by analytic continuation.

The Green function $G(ij, \tau)$ can be expanded with respect to the perturbation $\mathcal{H}_1(\tau)$ in a systematic way (see for instance Abrikosov *et al* 1963). This expansion involves the calculation of terms like $\langle T_\tau S_i(\tau) S_j(0) \mathcal{H}_1(\tau_1)\mathcal{H}_1(\tau_2)\ldots\rangle_0$, where the average is with respect to the non-interacting $\mathcal{H}_0$ as denoted by the suffix 0. This averaging leaves only the products in which all the spin operators belong to the same site, and we define ($p = 2, 4, \ldots$) the pth order semi-invariant

$$S^{(p)} = \int_0^\beta \mathrm{d}\tau_1 \ldots \int_0^\beta \mathrm{d}\tau_p \left\langle T_\tau\left(\prod_{j=1}^{p} S(\tau_j)\right)\right\rangle_0 \prod_{j=1}^{p}\exp(\mathrm{i}\omega_{n,j}\tau_j). \tag{4a}$$

The spin cumulants or the (reduced) semi-invariants are then defined as

$$\Gamma_p(\mathrm{i}\omega_{n,1}, \ldots, \mathrm{i}\omega_{n,p-1}) = S^{(p)} - \sum \Gamma_{p1}\Gamma_{p2}\ldots\Gamma_{pm} \tag{4b}$$

where the sum is over all possible products of two or more semi-invariants for which $p_1 + p_2 \ldots + p_m = p$. The most important reason for considering Γ_p rather than $S^{(p)}$ is that it makes it possible to reduce the expansion of $G(ij, \tau)$ so that it only contains linked diagrams. We notice that because of time-invariance $S^{(p)}$ vanishes unless $\omega_{n,p} = -(\omega_{n,1} + \ldots + \omega_{n,p-1})$, so Γ_p only depends on $p - 1$ frequency variables. Furthermore, if the spin operators were Bose operators then all the cumulants except $\Gamma_2(\mathrm{i}\omega_n)$ would vanish. We shall first consider this case, for which the diagrammatic expansion reduces to the one analysed by Schmidt (1974), and Lage and Stinchcombe (1977), using the method of Yonezawa (1968). We shall not discuss this procedure here, but refer to the original papers. Instead we shall consider an alternative way leading to the CPA result.

The single sites are considered to possess different properties depending on whether $c_j = 1$ or 0, but they are assumed to interact with a medium which is the same for all the sites, and this configurationally averaged medium is established in a self-consistent fashion. This means that $G(ij, \tau)$, in the case $i = j$, is approximated by

$$G(jj, \tau) \simeq c_j^2 g(\tau) = c_j g(\tau) \tag{5a}$$

where

$$g(\tau) = \langle T_\tau S_j(\tau) S_j(0) \rangle_{\text{(effective medium)}} \tag{5b}$$

is independent of the local disorder, i.e. the site index. In figure 1 we present the CPA diagrams arranged in a way which allows us to consider the single sites. In these diagrams we operate with two kinds of renormalised, frequency-dependent, interaction lines (the wavy double lines) defined in terms of $\tilde{J}(ij, \mathrm{i}\omega_n)$. This interaction is a sum of the infinite series of interaction-chain diagrams connecting the sites i and j. In order to prevent the different interaction-chain diagrams from contributing more than once in the final result, we include in $\tilde{J}(ij, \mathrm{i}\omega_n)$ only those diagrams where the ith site appears at the beginning and the jth site at the end of the interaction chains, but nowhere in between. The wavy double line without the cross is the interaction $K(\mathrm{i}\omega_n) = \delta_{ij}\langle \tilde{J}(ij, \mathrm{i}\omega_n)\rangle$. This connects the site j with itself once via the effective medium established as the configurational average of $\tilde{J}(ij, \mathrm{i}\omega_n)$. In terms of $K(\mathrm{i}\omega_n)$ the single-site Green function, (5), can be expressed in a simple series of diagrams where the jth site appears once, twice etc, or

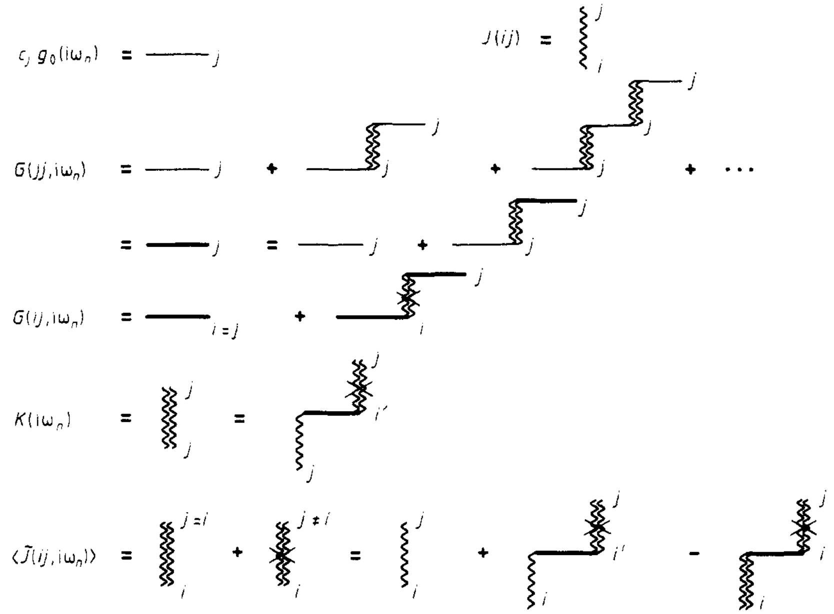

Figure 1. The diagrammatic representation of the CPA Green function, $G(ij, \mathrm{i}\omega_n)$, in co-ordinate space for a dilute system. The internal site index i' implies a summation over all sites averaged over all configurations (c_i is replaced by the average value c).

referring to figure 1:

$$g(\mathrm{i}\omega_n)=\frac{g_0(\mathrm{i}\omega_n)}{1-K(\mathrm{i}\omega_n)g_0(\mathrm{i}\omega_n)}\qquad g_0(\mathrm{i}\omega_n)=\Gamma_2(\mathrm{i}\omega_n)\tag{6}$$

where $g_0(\mathrm{i}\omega_n)$ is the non-interacting Green function. Corresponding to (5) the sites i and j are considered to be coupled via the effective medium, and $G(ij,\mathrm{i}\omega_n)$, $i\neq j$, is determined by $K(ij,\mathrm{i}\omega_n)=(1-\delta_{ij})\langle\bar{J}(ij,\mathrm{i}\omega_n)\rangle$, denoted by a cross on the wavy line, as shown in figure 1. Note that the occurrence of $c_i g(\mathrm{i}\omega_n)$ and $c_j g(\mathrm{i}\omega_n)$ in the diagram determining $G(ij,\mathrm{i}\omega_n)$ accounts for the interaction-chain diagrams where the ith or jth sites appear more than once. $\langle\bar{J}(ij,\mathrm{i}\omega_n)\rangle$ is determined self-consistently in terms of the bare interaction and the single-site Green function as shown in the lower part of figure 1. The third diagram of $\langle\bar{J}(ij,\mathrm{i}\omega_n)\rangle$ has to be subtracted because the site i appears between the sites i' and j in the second diagram. Here we utilise directly equation (5) as $c_{i'}g(\mathrm{i}\omega_n)$, appearing when the chains involving the intermediate site i' are summed, is considered to be determined by the effective medium without special reference to the actual neighbours i or j. The equations corresponding to the diagrams are straightforwardly solved after transforming to $\boldsymbol{q}$-space, and defining

$$D(c;\boldsymbol{q},\mathrm{i}\omega_n)=\frac{1}{1-[J(\boldsymbol{q})-K(\mathrm{i}\omega_n)]cg(\mathrm{i}\omega_n)}.\tag{7}$$

$K(\mathrm{i}\omega_n)$ is then determined by the condition that

$$\frac{1}{N}\sum_{\boldsymbol{q}}D(\boldsymbol{q},\mathrm{i}\omega_n)=1\tag{8a}$$

which may be written

$$K(\mathrm{i}\omega_n)=\frac{1}{N}\sum_{\boldsymbol{q}}J(\boldsymbol{q})D(\boldsymbol{q},i\omega_n)\tag{8b}$$

whereas

$$K(\boldsymbol{q},\mathrm{i}\omega_n)=[J(\boldsymbol{q})-K(\mathrm{i}\omega_n)]D(\boldsymbol{q},\mathrm{i}\omega_n).\tag{8c}$$

$(1/N)\Sigma_{\boldsymbol{q}}K(\boldsymbol{q},\mathrm{i}\omega_n)\equiv0$ is the T-matrix for the effective single-site Green function, which vanishes in accordance with an alternative formulation of the CPA (see Elliott *et al* 1974). Finally, we get

$$\langle G(c;\boldsymbol{q},\mathrm{i}\omega_n)\rangle=cg(\mathrm{i}\omega_n)D(\boldsymbol{q},\mathrm{i}\omega_n)\tag{9}$$

which, within the RPA, equation (6), may be written in the closed form

$$[\langle G(c;\boldsymbol{q},\mathrm{i}\omega_n)\rangle]^{-1}=[G(c=1;\boldsymbol{q},\mathrm{i}\omega_n)]^{-1}+(1-c)\left[\frac{1}{n}\sum_{\boldsymbol{q}'}\langle G(c;\boldsymbol{q}',\mathrm{i}\omega_n)\rangle\right]^{-1}.\tag{10}$$

This expression is the same as that derived from the self-contained CPA cumulant expansion of Yonezawa (1968); thus the two procedures are equivalent.

In the concentrated limit, $c=1$, the result above reduces to the RPA

$$G(c=1;\boldsymbol{q},\mathrm{i}\omega_n)=\frac{g_0(\mathrm{i}\omega_n)}{1-J(\boldsymbol{q})g_0(\mathrm{i}\omega_n)}\tag{11}$$

In the systematic expansion in powers of $1/Z$ the chain diagrams are classified according to the number of independent momentum labels which appear and which are eventually summed over (see e.g. Stinchcombe 1973). The RPA Green function is the result obtained to zeroth order of $1/Z$. If the fourth-order semi-invariants Γ_4 are non-zero, they introduce two extra interaction lines in the chains. To first order in $1/Z$, only the diagrams in which the two extra lines merge into one are included. These diagrams contain one extra interaction line which starts and terminates at the same site. Therefore in order $1/Z$ the single sites can be considered separately, allowing the diagrams to be arranged in the same way as in figure 1. The only modification is the inclusion of the fourth-order cumulant in the single-site Green function as illustrated in figure 2(*a*):

$$g_1(\mathrm{i}\omega_n) = \Gamma_2(\mathrm{i}\omega_n) + \sum_{n1,n2,n3} \Gamma_4(\mathrm{i}\omega_n, \mathrm{i}\omega_{n1}, \mathrm{i}\omega_{n2})\, \tilde{K}(\mathrm{i}\omega_{n3}) \qquad (12a)$$

and

$$g(\mathrm{i}\omega_n) = \bar{g}(\mathrm{i}\omega_n) = \frac{g_1(\mathrm{i}\omega_n)}{1 - K(\mathrm{i}\omega_n)\, g_1(\mathrm{i}\omega_n)} \qquad (12b)$$

where the unrestricted interaction is

$$\tilde{K}(\mathrm{i}\omega_n) = K(\mathrm{i}\omega_n)\big(1 + \bar{g}(\mathrm{i}\omega_n)\, K(\mathrm{i}\omega_n)\big). \qquad (12c)$$

The remaining diagrams in figure 1 are unaltered and the result is the same as above with $g(\mathrm{i}\omega_n)$ determined by equation (12) instead of equation (6). When $c = 1$ these equations reduce to equation (11) with $g_0(\mathrm{i}\omega_n)$ replaced by $g_1(\mathrm{i}\omega_n)$ and

$$\tilde{K}(c = 1; \mathrm{i}\omega_n) = \frac{1}{N} \sum_{q} \frac{J(q)}{1 - J(q)\, g_1(\mathrm{i}\omega_n)} \qquad (13)$$

which is the usual way of presenting the $1/Z$ result. The considerations above are equally valid when c is less than 1, and equations (7)–(9) together with equation (12) determine the average two-site Green function, when the CPA is combined with the $1/Z$ expansion to first order in $1/Z$.

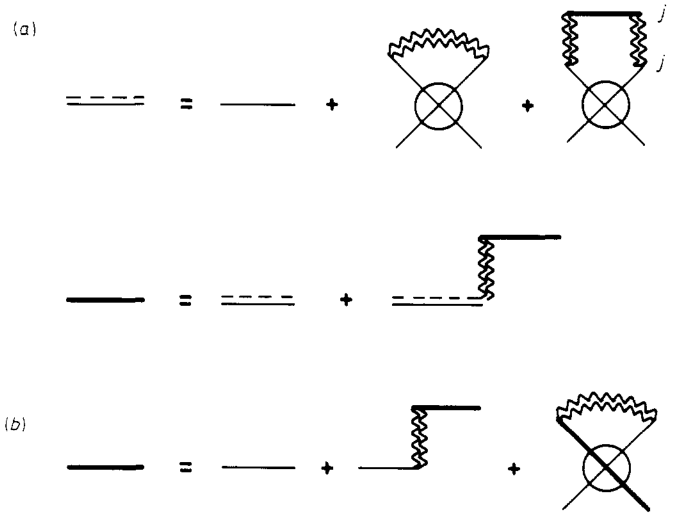

Figure 2. The single-site Green function, $g(\mathrm{i}\omega_n)$, to first order in $1/Z$. The circles represent the fourth-order cumulant Γ_4. (*a*) shows $g_1(\mathrm{i}\omega_n)$ and $g(\mathrm{i}\omega_n) = \bar{g}(\mathrm{i}\omega_n)$, which includes effects of Γ_2 and Γ_4 to infinite order, equation (12). (*b*) gives $g(\mathrm{i}\omega_n)$ in the approximation where (most) terms in $1/g(\mathrm{i}\omega_n)$ of higher order than $1/Z$ are neglected, equation (14).

The single-site Green function $g(\mathrm{i}\omega_n)$ does, however, contain terms of higher order than $1/Z$. In order to include these terms one makes implicitly the assumption that $\Gamma_p \approx 0$ for $p > 4$, corresponding to a decoupling of $S^{(p)}$ into products of the lower-order semi-invariants (see equation (4)). In the next section we consider specifically $S = 1$, and we find that this decoupling is not a good approximation. In the cumulant expansion of $g(\mathrm{i}\omega_n)$ the semi-invariant Γ_6 is multiplied by a factor of the order $(1/Z)^2$ and is therefore of minor importance, but the inaccuracies are present in every term of the infinite series and might sum to be of decisive importance. In the next section we consider situations where the approximate expression for $g(\mathrm{i}\omega_n)$ given by equation (12) is unreliable or even useless. In order to circumvent this difficulty we shall consider instead the single-site Green function shown in figure 2(b), which we may express formally as

$$\begin{aligned} g(\mathrm{i}\omega_n) = {} & \Gamma_2(\mathrm{i}\omega_n) + \Gamma_2(\mathrm{i}\omega_n)\,K(\mathrm{i}\omega_n)\,g(\mathrm{i}\omega_n) \\ & + \sum_{n1,n2,n3} \Gamma_4(\mathrm{i}\omega_n, \mathrm{i}\omega_{n1}, \mathrm{i}\omega_{n2})\,K(\mathrm{i}\omega_{n3})\,g(\mathrm{i}\omega_m)/\Gamma_2(\mathrm{i}\omega_m) \end{aligned} \tag{14}$$

where m might be equal to n or $n3$ depending on the term considered. Although this seems to introduce some arbitrariness, it turns out to be possible to give a unique prescription for the choice of m in the $S = 1$ case. The two expressions for the single-site Green function, (12) and (14), agree within the accuracy with which the calculation is actually performed, namely to first order in $1/Z$, and they both lead to the RPA if Γ_4 vanishes. However, the replacement of $\bar{g}(\mathrm{i}\omega_n)$, equation (12), with $g(\mathrm{i}\omega_\mathrm{n})$, equation (14), implies that second and higher-order changes due to Γ_4 in the position of the poles of the Green functions are neglected.

In the next section we discuss the results above in the case $S = 1$, but before doing that we want to extend the CPA-calculation to the case where both the two kinds of atoms, type 1 and 2, have non-zero spins, S_1 and S_2. The interaction in equation (1) is replaced by a 2×2 matrix, $J_{\alpha\beta}(ij)$, where e.g. $J_{12}(ij)$ describes the coupling of $c_i S_{1,i}$ with $(1 - c_j) S_{2,j}$. The effective single-site approach is straightforwardly generalised to this case as shown by the diagrams in figure 3. The two single-site Green functions, $g_{(1)}(\mathrm{i}\omega_n)$ and $g_{(2)}(\mathrm{i}\omega_n)$, are determined as before in terms of the restricted self-interactions $K_{11}(\mathrm{i}\omega_n)$ and $K_{22}(\mathrm{i}\omega_n)$ respectively. We define

$$I_{\alpha\beta}(\boldsymbol{q}, \mathrm{i}\omega_n) = J_{\alpha\beta}(\boldsymbol{q}) - K_{\alpha\beta}(\mathrm{i}\omega_n) \tag{15}$$

and the determinants of the 2×2 matrices like, for instance,

$$|J(\boldsymbol{q})| = J_{11}(\boldsymbol{q}) J_{22}(\boldsymbol{q}) - J_{12}^2(\boldsymbol{q}). \tag{16}$$

Furthermore we define

$$\begin{aligned} D(c; \boldsymbol{q}, \mathrm{i}\omega_n) = {} & [1 - I_{11}(\boldsymbol{q}, \mathrm{i}\omega_n)\,G_1(\mathrm{i}\omega_n) - I_{22}(\boldsymbol{q}, \mathrm{i}\omega_n)\,G_2(\mathrm{i}\omega_n) \\ & + |I(\boldsymbol{q}, \mathrm{i}\omega_n)|\,G_1(\mathrm{i}\omega_n)\,G_2(\mathrm{i}\omega_n)]^{-1} \end{aligned} \tag{17}$$

where $G_1(\mathrm{i}\omega_n) = c g_{(1)}(\mathrm{i}\omega_n)$ and $G_2(\mathrm{i}\omega_n) = (1 - c) g_{(2)}(\mathrm{i}\omega_n)$. The 2×2 matrix of the averaged two-site Green functions is then found to be

$$\langle G_{11}(\boldsymbol{q}, \mathrm{i}\omega_n)\rangle = G_1(\mathrm{i}\omega_n)[1 - I_{22}(\boldsymbol{q}, \mathrm{i}\omega_n)\,G_2(\mathrm{i}\omega_n)]\,D(\boldsymbol{q}, \mathrm{i}\omega_n) \tag{18a}$$

with a similar expression for $\langle G_{22}(\boldsymbol{q}, \mathrm{i}\omega_n)\rangle$, whereas

$$\langle G_{12}(\boldsymbol{q}, \mathrm{i}\omega_n)\rangle = G_1(\mathrm{i}\omega_n)\,G_2(\mathrm{i}\omega_n)\,I_{12}(\boldsymbol{q}, \mathrm{i}\omega_n)\,D(\boldsymbol{q}, \mathrm{i}\omega_n). \tag{18b}$$

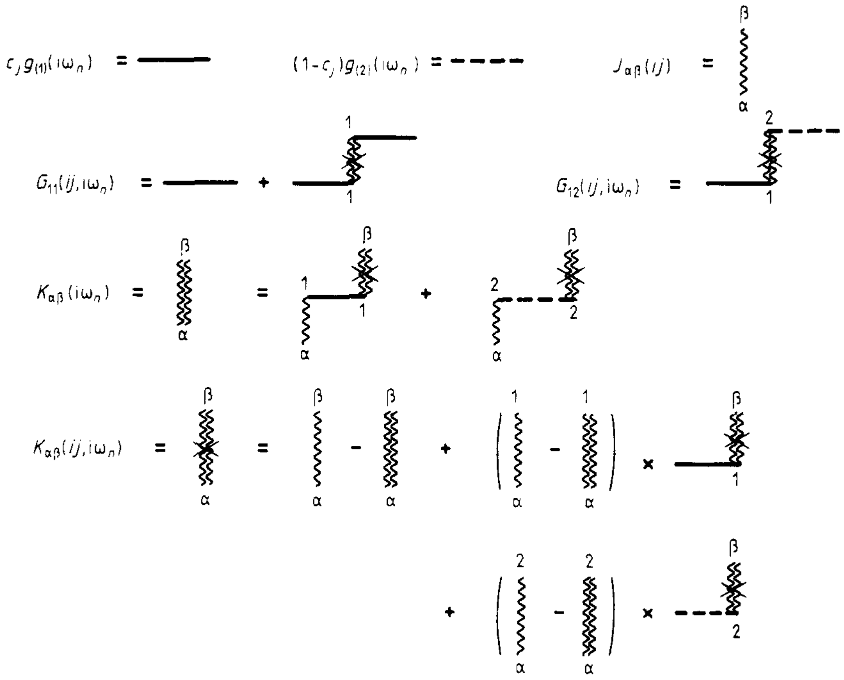

Figure 3. The CPA Green function for a binary alloy system. α and β denote the two kinds of sites, 1 or 2. The site indices which are suppressed appear in the same way as in figure 1.

The only problem left is the determination of closed expressions for $K_{\alpha\beta}(i\omega_n)$. In order to obtain these it is more convenient to use the conditions $(1/N)\sum_q K_{\alpha\beta}(\boldsymbol{q}, i\omega_n) \equiv 0$, rather than directly using the expressions given by the diagram in figure 3, and we find

$$K_{\alpha\beta}(i\omega_n) = \frac{1}{B(i\omega_n)}\left[A_{\alpha\beta}(i\omega_n) + \delta_{\alpha\beta}\frac{1 - B(i\omega_n)}{G_\alpha(i\omega_n)}\right] \tag{19a}$$

where

$$A_{\alpha\beta}(i\omega_n) = \frac{1}{N}\sum_q J_{\alpha\beta}(\boldsymbol{q})D(\boldsymbol{q}, i\omega_n) \tag{19b}$$

and

$$\begin{aligned} B(i\omega_n) = (1/N)\sum_q D(\boldsymbol{q}, i\omega_n) &= [1 - G_1(\omega)G_2(\omega)\,|A(i\omega_n)|\,] \\ &\times [1 - G_1(\omega)G_2(\omega)(1/N)\sum_q |J(\boldsymbol{q})|\,D(\boldsymbol{q}, i\omega_n)]^{-1}. \end{aligned} \tag{19c}$$

In a comparison with the CPA result obtained in the dilute case we notice that $B(i\omega_n) \neq 1$ represents a non-trivial modification. In cases where the geometrical-mean approximation is valid, i.e. $J_{12}(\boldsymbol{q}) = \gamma J_{11}(\boldsymbol{q})$ and $J_{22}(\boldsymbol{q}) = \gamma^2 J_{11}(\boldsymbol{q})$, all the determinants vanish, implying $B(i\omega_n) = 1$. This approximation might be useful for analysing binary rare-earth systems (Jensen 1979). If, further, the single-site Green functions are represented by their RPA values, the result becomes equivalent to the CPA in the case of a phonon system with diagonal disorder (in which case $1/g_0(i\omega_n) \propto \omega_E^2 - (i\omega_n)^2$, where ω_E, the Einstein oscillator frequency, is the same for the two kinds of atom).

3. The anisotropic $S = 1$ paramagnet

The theory developed in the preceding section is general. In order to apply it to a particular system, the only quantity which is needed is the Green function for a single site interacting with an effective medium described by $K(\mathrm{i}\omega_n)$. Here we shall discuss the case $S = 1$, and the single-site Hamiltonian is assumed to be

$$\mathcal{H}_0 = \Delta S_z^2. \tag{20}$$

We shall mainly be interested in the case where Δ is positive, but the calculation is equally valid if Δ is negative. In the easy-planar case of $\Delta > 0$, the ground state is the singlet $|0\rangle$ and the excited doublet $|\pm 1\rangle$ is separated from the singlet by the energy Δ. The single-site Green function has three components, of which two are degenerate. In the non-interacting case these are

$$g_0^{xx}(\mathrm{i}\omega_n) = g_0^{yy}(\mathrm{i}\omega_n) = g_0(\mathrm{i}\omega_n) = \frac{2\Delta n_{01}}{\Delta^2 - (\mathrm{i}\omega_n)^2} \tag{21a}$$

and

$$g_0^{zz}(\mathrm{i}\omega_n) = 2\beta n_1 \delta_{n,0} \tag{21b}$$

where we have introduced the population factors

$$n_0 = (1 + 2\mathrm{e}^{-\beta\Delta})^{-1} \qquad n_1 = n_0 \exp(-\beta\Delta_{n_0}) \tag{21c}$$

and $n_{01} \equiv n_0 - n_1$. In this section we shall be concerned with crystals where only the one type of atoms has a non-zero spin. The Green functions are then diagonal with respect to the Cartesian components, with three components for the interaction, $K_x(\mathrm{i}\omega_n) = K_y(\mathrm{i}\omega_n)$, and $K_z(\mathrm{i}\omega_n)$. In order to include the interaction to leading order we need to calculate the fourth-order semi-invariants multiplied by these interaction components. This can be carried out in a straightforward manner with the use of the Wick-like theorem derived by Care and Tucker (1977). Applying this and introducing the modifications indicated by equation (14) we get

$$\begin{aligned} g^{xx}(\mathrm{i}\omega_n) = {} & g_0(\mathrm{i}\omega_m) + g_0(\mathrm{i}\omega_n) K_x(\mathrm{i}\omega_n) g^{xx}(\mathrm{i}\omega_n) \\ & + [(1/n_{01}^2) g^{xx}(\mathrm{i}\omega_n) \{ n_3 g_0(\mathrm{i}\omega_n) K_x(\mathrm{i}\omega_n) + n_1 u_0(\mathrm{i}\omega_n) K_y(\mathrm{i}\omega_n) \\ & - g_0(\mathrm{i}\omega_n) \Omega_1^x - \tfrac{1}{2}(u_0(\mathrm{i}\omega_n) - \tfrac{3}{2}\beta n_1 n_{01}) \Omega_1^y + u_1(\mathrm{i}\omega_n) K_z(\mathrm{i}0) \} \\ & - (\Omega_2^x + \tfrac{1}{2}\Omega_2^y + \tfrac{1}{2}\Omega_2^z) g_0(\mathrm{i}\omega_n)] \end{aligned} \tag{22a}$$

with $n_3 \equiv n_0 + n_1 - n_{01}^2$ and we define

$$u_0(\mathrm{i}\omega_n) = \frac{1}{2\Delta^2} \{\Delta^2 + (\mathrm{i}\omega_n)^2\} g_0(\mathrm{i}\omega_n) \tag{22b}$$

and

$$u_1(\mathrm{i}\omega_n) = \frac{1}{\beta} g_0^2(i\omega_n) - \left(n_1 + \frac{3n_{01}}{2\beta\Delta}\right) g_0(\mathrm{i}\omega_n) - \left(\frac{1}{2} + \frac{3}{4} n_{01} - \frac{1}{\beta\Delta}\right) \beta n_1 n_{01}. \tag{23}$$

Furthermore we have introduced the frequency sums ($\alpha = x, y$):

$$\begin{aligned}\Omega_1^\alpha &= \frac{1}{\beta}\sum_{n'} g_0(\mathrm{i}\omega_{n'})\,K_\alpha(\mathrm{i}\omega_{n'}) \\ &= (n_0+n_1)\operatorname{Re}[K_\alpha(\Delta)] + \frac{1}{\pi}\int_0^\infty g_0(\omega)\operatorname{Im}[K_\alpha(\omega+\mathrm{i}0)]\coth(\beta\omega/2)\,\mathrm{d}\omega\end{aligned} \tag{24a}$$

(Re and Im denote the real and imaginary parts)

$$\Omega_2^\alpha = \frac{1}{\beta}\sum_{n'}\frac{1}{n_{01}^2}\{u_0(\mathrm{i}\omega_{n'}) - \tfrac{1}{2}\beta n_3\}g^{\alpha\alpha}(\mathrm{i}\omega_{n'})\,K_\alpha(\mathrm{i}\omega_{n'}) \tag{24b}$$

and

$$\Omega_2^z = \tfrac{1}{4}g^{zz}(\mathrm{i}0)\,K_z(\mathrm{i}0). \tag{24c}$$

To first order in $1/Z$ there is a minor inelastic contribution to $K_z(\mathrm{i}\omega_n)$, which we have disregarded in equation (22) as it only affects $g^{xx}(\mathrm{i}\omega_n)$ in the order $(1/Z)^2$. $g^{yy}(\mathrm{i}\omega_n)$ is determined by the same expression with x and y interchanged, whereas

$$\begin{aligned}g^{zz}(\mathrm{i}0) = 2n_1\beta + 2n_1\beta g^{zz}(\mathrm{i}0)\,K_z(\mathrm{i}0) \\ &+ \Big[g^{zz}(\mathrm{i}0)\{\tfrac{1}{2}\beta(1-6n_1)\,K_z(\mathrm{i}0) - \tfrac{1}{8}\beta(\Omega_1^x + \Omega_1^y)\} \\ &+ \frac{1}{n_{01}^2}\sum_{n'} u_1(\mathrm{i}\omega_{n'})\{g^{xx}(\mathrm{i}\omega_{n'})\,K_x(\mathrm{i}\omega_{n'}) + g^{yy}(\mathrm{i}\omega_{n'})\,K_y(\mathrm{i}\omega_{n'})\}\Big].\end{aligned} \tag{25}$$

As mentioned above, the interactions $K_\alpha(\mathrm{i}\omega_n)$ introduce an inelastic term in $g^{zz}(\mathrm{i}\omega_n)$ which we shall not be concerned with here.

According to equation (14) we have replaced one non-interacting Green function in every term arising from the fourth-order semi-invariants, collected in the square brackets of equations (22*a*) and (25), with a final single-site Green function. The most important substitution performed in equation (22*a*) is that wherever $g_0^2(\mathrm{i}\omega_m)$ or $g_0(\mathrm{i}\omega_m)u_0(\mathrm{i}\omega_m)$ occur in the original expression for Γ_4, one factor $g_0(\mathrm{i}\omega_m)$ has been replaced by $g^{\alpha\alpha}(\mathrm{i}\omega_m)$. The substitution, in the few minor terms left, has been performed so that the final $g^{xx}(\mathrm{i}\omega_n)$ behaves properly in both the limits $\beta \to 0$ and $\beta \to \infty$, which demands that the term $\frac{1}{2}\beta n_3$ is subtracted from $u_0(\mathrm{i}\omega_{n'})$ in (23*b*). $g^{zz}(\mathrm{i}0)$ is established using the same procedure or by a consideration of the limit $\Delta \to 0$. If these substitutions were not performed, as for instance in $\tilde{g}(\mathrm{i}\omega_n)$ based on equation (12), the Ω_1^α terms would lead to an improper response for $\mathrm{i}\omega_n$ close to Δ because of the singular behaviour of $g_0^2(\mathrm{i}\omega_n)$. The modifications introduced by (14) mean that we only include the contributions from Γ_4 which are linear in $K_\alpha(\mathrm{i}\omega_m)$ or $1/Z$ in an expansion of the inverse Green functions. This statement is strictly obeyed only in the limit $\Delta \to 0$; in other cases the Ω_2 terms still give rise to higher-order terms in $1/g(\mathrm{i}\omega_n)$. Apart from these minor terms we only include the leading-order shifts of the poles in the Green functions, but the procedure accounts simultaneously for the corresponding modifications of the oscillator strengths, which allows the maintenance of self-consistency.

The systematic procedure above leaves one improper term, namely the first term in $u_1(\mathrm{i}\omega_n)$, equation (23), which is still proportional to $g_0^2(\mathrm{i}\omega_n)$. This term cannot be neglected at zero frequency, as $u_1(\mathrm{i}0)$ would then diverge in the limit $\Delta \to 0$, but it has to be modified at finite frequencies. We shall not try to guess the kind of modifications expected in higher order, but we simply replace $u_1(\mathrm{i}\omega_n)$ occurring in (22*a*) and (25) with

$$u_1(\mathrm{i}\omega_n) = \frac{2n_{01}}{\beta\Delta} g_0(\mathrm{i}\omega_n) - \left(n_1 + \frac{3n_{01}}{2\beta\Delta}\right) g_0(\mathrm{i}\omega_n) - \left(\frac{1}{2} + \frac{3}{4}n_{01} - \frac{1}{\beta\Delta}\right) \beta n_1 n_{01} \tag{22c}$$

We have analysed the final two-site Green function, which is obtained by inserting equations (22) or (25) in (9), in the case of an FCC lattice, considering various values of c, Δ and β. The response functions, $\mathrm{Im}[g^{xx}(\omega + \mathrm{i}0)]$ and $\mathrm{Im}[\langle G(c; \boldsymbol{q}, \omega + \mathrm{i}0)\rangle]$, are found to behave properly (being positive for $\omega > 0$). At low temperatures, $\beta\Delta \gtrsim 2$, the Green function is similar to the one obtained in the RPA–CPA case, except that the RPA parameters are replaced by effective ones. In this limit $K_z(\mathrm{i}0)$ can be neglected in equation (22), and $G(c = 1; \boldsymbol{q}, \omega)$ is found to be nearly the same as deduced from the equations of motion and applied to Pr metal (Jensen 1982). For instance at $T = 0$ the factor $\bar{c} = (1 - \Omega_2^x - \frac{1}{2}\Omega_2^y)$ is equal to the self-consistent value of n_{01}, appearing in the equations of motion, except for higher-order corrections. In the present context this factor corresponds to the presence of non-magnetic impurities with concentration $1 - \bar{c}$ (in the FCC case $0.95 < \bar{c} < 1$), and it leads to a finite linewidth of the excitations. This prediction is consistent with the observation that spin fluctuations are present in the paramagnetic case at $T = 0$, because the product of the single-site states, $|0\rangle$, is not the true many-body ground state. The most important contributions to the temperature dependence of the linewidth, when $\beta\Delta \gtrsim 2$, is the first term in the square bracket of equation (22*a*). This has been discussed in detail by Bak (1975) in an application to Pr metal. The present result does not modify this discussion very much, but we may add that, if the system is diluted, the effect of the static disorder on the linewidth is equivalent to an increase in temperature, with the linewidth being proportional to approximately $(1 - cn_{01}^2)$. The consequences of the theory at non-zero frequencies will be discussed in more detail elsewhere, in connection with its application to the Pr–Nd alloy system. Here we shall examine more closely the predictions of the ground-state properties.

In the limit $\Delta \rightarrow 0$, the frequency sums, equation (24), can be performed at once as they only contain the terms at zero frequency, and we obtain from equation (22), with $\mathrm{i}\omega_n$ replaced by ω,

$$g^{xx}(0) = \frac{2}{3}\beta \frac{1 - (1/8)\{g^{yy}(0)K_y(0) + g^{zz}(0)K_z(0)\}}{1 - (\beta/12)\{2K_x(0) - K_y(0) - K_z(0)\}} \tag{26}$$

which is the same as given by $g^{zz}(0)$, equation (25), when x and z are interchanged. In the three different models where one, two, or the three components of $K(0)$ are non-zero and equal, the inverse susceptibilities are

$$\frac{1}{g(0)} = \frac{3}{2\beta} + \begin{cases} -\frac{1}{4}K(0) & \text{Ising} \\ 0 & XY \\ +\frac{1}{4}K(0) & \text{Heisenberg} \end{cases} \tag{27}$$

determined by equations (22) or (26) to first order in $1/Z$. Now in this limit the spin operators in the interaction representation no longer depend on τ. Furthermore, in the Ising model the only spin operator which occurs in the cumulant expansion is S_z, which commutes with itself. This means that in the effective-medium approximation where the single site is perturbed by

$$\mathcal{H}_1(\tau_1, \tau_2) = -\tfrac{1}{2} K(\tau_1 - \tau_2) S_z(\tau_1) S_z(\tau_2) \tag{28a}$$

the cumulant expansion for $g_1(0)$ can be summed exactly, as the perturbation becomes the equivalent to a static anisotropy

$$\mathcal{H}_{\mathrm{a}} = -\tfrac{1}{2}K(0)S_z^2 \tag{28b}$$

in the Hamiltonian, or

$$g_{\mathrm{I}}(0)_{\mathrm{ex}} = 2\beta/[2 + \exp(-\tfrac{1}{2}\beta K(0))] \tag{29}$$

when $S = 1$. The use of $g_{\mathrm{I}}(0)_{\mathrm{ex}}$ instead of the other approximate expressions for $g_{\mathrm{I}}(0)$ gives the most accurate effective-medium theory for the Ising model. The results obtained in this fashion coincide with that of Lines' somewhat less rigorous correlated-effective-field theory (1974, 1975). In his theory the interaction (28*a*) is in general considered as simulated by an effective anisotropy like (28*b*). However, because of the τ-ordering this does not hold true when other non-commuting spin operators, S_x or S_y, are added to the interaction Hamiltonian, or if e.g. the $|\pm 1\rangle$ level is split so that the interaction becomes retarded, as described by the τ dependence in (28*a*), which effect is not included in (28*b*). In the *XY* and Heisenberg models the presence of non-commuting operators in the spin-traces introduces essential complications but, by utilising the τ independence of the operators, it is rather straightforward to extend the expansion of $g(0)$ to second (or higher) order in $K(0)$ or $1/Z$, and we get

$$g_{XY}(0) = \tfrac{2}{3}\beta \frac{1 + (2/3!!)x + (3/5!!)x^2 + (4/7!!)x^3 + \ldots}{1 + \tfrac{2}{3}(x + (1/3!!)x^2 + (1/5!!)x^3 + \ldots)} \tag{30a}$$

with $x = \beta K(0)$ and

$$g_{\mathrm{H}}(0) = \tfrac{2}{3}\beta \frac{\tfrac{2}{3}((3!!/2!) + 2(5!!/4!)x + 3(7!!/6!)x^2 + \ldots)}{1 + \tfrac{2}{3}((3!!/2!)x + (5!!/4!)x^2 + (7!!/6!)x^3 + \ldots)}. \tag{30b}$$

These expressions, (30*a*) and (30*b*), are calculated under the assumption of a static effective medium, $K(\mathrm{i}\omega_n) = K(\mathrm{i}0)\,\delta_{n,0}$. Nevertheless, $g_{\mathrm{H}}(\tau)$ is going to depend on τ:

$$g_{\mathrm{H}}(\tau) = \tfrac{2}{3} - \tfrac{1}{3}\beta K(0)\{1 - \tau^2/\beta^2 - (\beta - \tau)^2/\beta^2\} \tag{30c}$$

to the order $1/Z$, which result is consistent with the sum rule

$$\sum_{\alpha = x,y,z} g^{\alpha\alpha}(\tau = 0) = S(S+1) = 2$$

and with $g_{\mathrm{H}}(\omega = 0)$ as given by (30*b*) or (27). Equation (30*c*) indicates the presence of a diffusive peak in the response function of width $\sim 2[2K(0)/\beta]^{1/2}$. The τ-dependent terms in (30*c*) are going to generate inelastic contributions to $K(\mathrm{i}\omega_n)$ which will modify (30*b*) in the order $(1/Z)^2$. These contributions may be of some importance, quantitatively, when $\beta K(0) > 1$. But they should not change the qualitative behaviour indicated by (30*b*), and we shall not consider this modification in any further detail. Similar remarks apply to the *XY* model, equation (30*a*), whereas there are no modifications of $g_{\mathrm{I}}(0)$ in equation (29) as only the transverse components of the susceptibility tensor are going to depend on τ in the case of the Ising model.

In the theory of Lines, $g_{XY}(0)$ is determined by changing the sign of $K(0)$ in equation (28*b*), whereas $g_{\mathrm{H}}(0) = \tfrac{2}{3}\beta$. The RPA values of $g(0)$, equation (6), are the same in all three models, namely

$$g_{\mathrm{RPA}}(0) = \tfrac{2}{3}\beta/(1 - \tfrac{2}{3}\beta K(0)). \tag{31}$$

If we consider $K(0)$ to be fixed by some external conditions, then $g_{\mathrm{RPA}}(0)$ diverges at $1/\beta = \tfrac{2}{3}K(0)$, which divergence is not present in the exact case of equation (29). This

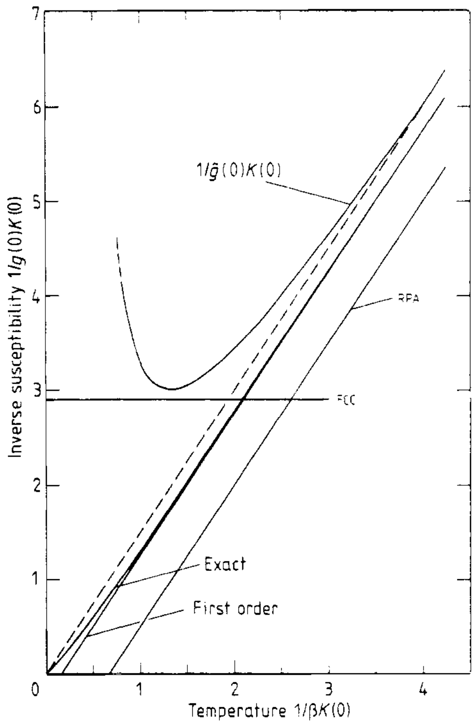

Figure 4. Comparison of the various expressions for the inverse single-site susceptibility of the Ising model. The interaction with the surroundings, $K(0)$, is considered to be a constant, and the broken line is the non-interacting inverse susceptibility. The horizontal line gives the critical value, $1/g(0)K(0) = c/\lambda$, in the FCC case when $c = 1$.

behaviour is non-physical as the system should be stable as long as self-consistency is neglected. If we finally include the single-site Green function, calculated to first-order in $(1/Z)$ as given by equation (12), in our comparison, then in the Ising case this is determined by

$$\tilde{g}_{\mathrm{I}}(0) = [\tfrac{2}{3}\beta - \tfrac{1}{3}\beta^2 K(0)(1 + \tilde{g}_{\mathrm{I}}(0)K(0))] \\ \times [1 - \tfrac{2}{3}\beta K(0) + \tfrac{1}{3}\beta^2 K^2(0)(1 + \tilde{g}_{\mathrm{I}}(0)K(0))]^{-1}. \qquad (32)$$

$\tilde{g}_{\mathrm{I}}(0)$ is closer to the exact value than $g_{\mathrm{RPA}}(0)$ at high temperatures but becomes negative for $1/\beta < \frac{1}{2}K(0)$. In figure 4 we compare the different approximations for $g_{\mathrm{I}}(0)$ when $K(0)$ is considered to be fixed. This comparison shows that the expressions $g_{\mathrm{RPA}}(0)$ and $\tilde{g}_{\mathrm{I}}(0)$ are hardly acceptable for $1/\beta < 2K(0)$, whereas the first-order approximation of equation (27) closely reproduces the exact behaviour down to $(1/\beta) \simeq 0.5\, K(0)$, and the temperature at which this susceptibility diverges, $1/\beta = \frac{1}{6}\, K(0)$, is reduced by a factor of four in comparison with $g_{\mathrm{RPA}}(0)$. The comparison shows consistently with equation (22) that the higher-order terms occurring in $\tilde{g}(0)$ are unreliable. Not even the terms of second order in $1/Z$ are of any use as they are a factor of ±40–50 times the corresponding terms in the exact expressions, in all the three models.

We cannot expect the assumption of an effective medium to be valid in the close neighbourhood of a second-order phase transition. Nevertheless, the critical temperatures predicted in this approximation are close to the values obtained by high-temperature expansion techniques. At the transition to a ferromagnetic phase

$$K(0) = \frac{\lambda}{1+\lambda} J(\mathbf{0}) \tag{33a}$$

where we define

$$\lambda \equiv \frac{1}{N} \sum_{q} \frac{J(q)}{J(\mathbf{0}) - J(q)} \tag{33b}$$

and the critical temperature, $T_c = 1/k_B\beta_0$, is determined as the temperature at which $G(\mathbf{0}, 0)$ diverges, i.e. by

$$cg(0)J(\mathbf{0}) = 1 + \lambda. \tag{34}$$

We shall limit ourselves to the case of an FCC crystal with only nearest-neighbour coupling where $\lambda = 0.34466$. In the Ising case, the use of the exact expression for $g(0)$, (29), predicts $1/\beta_0 = 0.534J(\mathbf{0})$ and $c = 1$, which is about 6% smaller than the value obtained from high-temperature expansions, $(1/\beta_0)(\text{HTE}) = 0.568J(\mathbf{0})$. The RPA value is $(1/\beta_0)(\text{RPA}) = \frac{2}{3}J(\mathbf{0})$, whereas $\bar{g}_I(0)$ does not produce a second-order transition in this system. The first-order result for $g(0)$, given by (27) when $\Delta = 0$ and determined by equation (22) when $\Delta \neq 0$, is estimated to be close to the exact value of the single-site Green function, and in figure 5 we show the critical temperature predicted in this

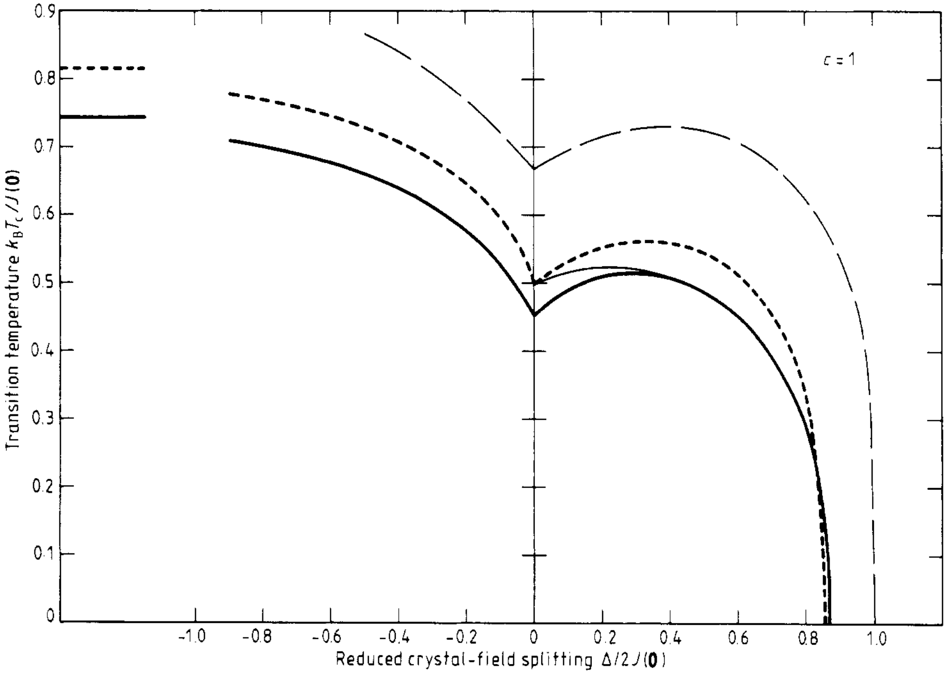

Figure 5. The critical temperature for the $S = 1$ anisotropic Heisenberg model in the FCC case, as a function of the anisotropy parameter, $\Delta/2J(\mathbf{0})$. The thin broken curve shows the RPA behaviour, and the thick broken curve is the estimate from high-temperature expansions. The full curve shows the prediction of the effective-medium theory with $g(i\omega_n)$ determined by equations (22)–(24). For comparison we also show the result obtained with an XY coupling for $\Delta \geqslant 0$, the thin unbroken line which joins the thick one for $\beta_0\Delta \leqslant 2$. The lines in the left side of the figure show the asymptotic behaviour when $\Delta \to -\infty$ (the RPA value is $k_BT_c/J(\mathbf{0}) = 1$) corresponding to an $S = \frac{1}{2}$ Ising model.

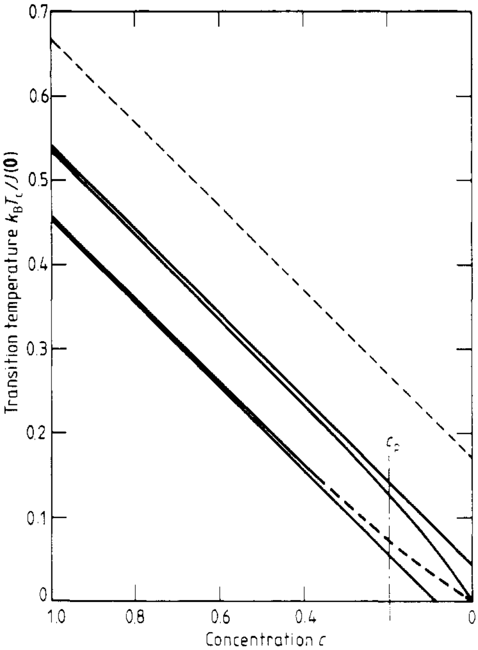

Figure 6. The critical temperature as a function of concentration in the FCC case. c_p denotes the percolation concentration. The broken curve is the RPA-CPA result. The full curves show the predictions of the effective-medium theory for the Ising model (the upper ones) and the Heisenberg model (the lower ones) using the first-order (the straight lines) and the exact single-site Green functions of the static medium.

approximation compared with the RPA and HTE values (Johnson and Wang 1981, Yang and Wang 1975 and references therein). The comparison of the XY and the Heisenberg models for $\Delta > 0$ shows that the S_z coupling is without importance for $\beta\Delta \gtrsim 2$. Furthermore we find that the system stays paramagnetic at $T = 0$ if the ratio $\Delta/2J(\mathbf{0})$ is larger than the critical value $r = 0.869 \pm 0.002$ which is close to the value of 0.858 predicted by HTE (Johnson and Wang 1981). The inelastic $1/Z$ contribution to $K_z(i\omega_n)$ neglected in the expression for $g^{xx}(i\omega_n)$ introduces a difference between the anisotropic Heisenberg and XY models in the $T = 0$ limit, as it reduces the critical ratio for the Heisenberg model, but only by about one tenth of a percent. This difference is proportional to $(1/Z)^2$ and cannot be trusted as long as other terms of this order are neglected. We remark that Yang and Wang (1975) have produced a result to first order in $1/Z$ which predicts a critical temperature in the easy-axis $S = 1$ case, in close agreement with HTE. However, we consider this to be a somewhat fortuitous coincidence, as they do not determine the effective medium in a fully self-consistent way but rely on an expansion in J. The comparison in figure 5 shows the same trend as already found in the Ising model, that the effective-medium approximation suppresses the critical temperature by 5–10%, which discrepancy should be considered as a genuine effect of the approximation. Comparing this number with the total modification of the MF/RPA behaviour induced by fluctuations, we can say that the effective-medium theory overestimates the renormalisation of the interactions by less than ~25%.

When the system is diluted, the critical temperature decreases roughly proportional to c. In figure 6 we compare the c dependences of the critical temperature of the Ising and Heisenberg models obtained by using either (27) or the exact single-site Green functions of the static medium (29) or (30b). In the Heisenberg case we have utilised the systematic behaviour of the terms calculated and given in (30*b*) for an extrapolation to $T = 0$ (indicated by the broken line in the figure). In figure 7 we show the critical ratio r, as a function of c, in the presence of a Heisenberg or an XY coupling, obtained from equations (22) and (24). The $1/Z$ contributions at $T = 0$, Ω_1^α and Ω_2^α, both vanish in the limit $c \rightarrow 0$, because the inelastic spectral density goes to zero. Therefore the CPA result approaches the result obtained in RPA, which is shown by the broken curve in the figure.

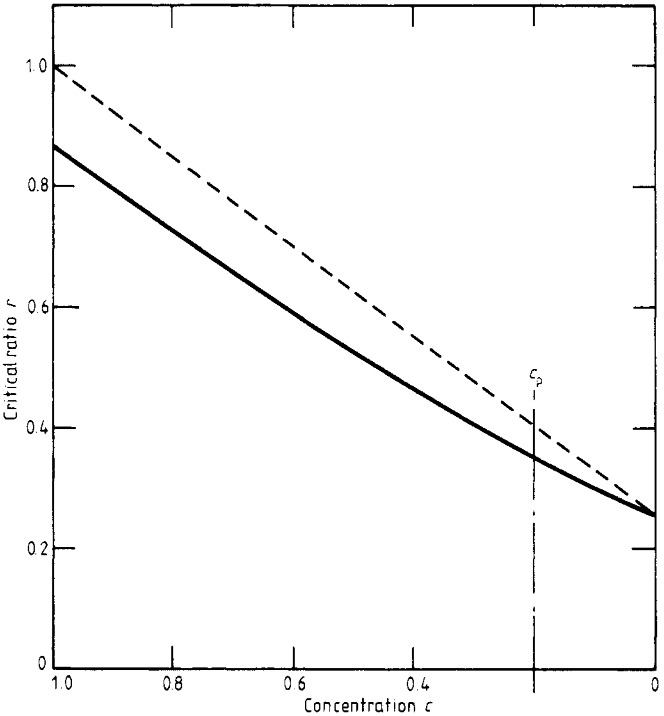

Figure 7. The critical ratio r as a function of concentration in the FCC case. The full curve is the prediction of the effective-medium theory using the first-order single-site Green function, which is compared with the RPA–CPA result shown by the broken curve. c_p is the percolation concentration.

The first-order results used for $g(0)$ at the lowest concentrations are unreliable, but this is the case also with the effective-medium approximation itself, which ceases to be valid because of percolation effects (Elliott *et al* 1974). In the present system the critical concentration c_p below which no long-range ordering can occur is calculated to lie between 0.195 and 0.199 (Essam 1972). Nevertheless, the spurious divergence of $g(0)$ occurring in the order $1/Z$, which leads to the finite critical ratio in the limit $c \rightarrow 0$, is inconvenient, as the results are distorted somewhat in the regimes where the effective-medium approximation is reasonable. However, the situation is, in general, much improved in comparison with the RPA–CPA results. If, for instance, the non-magnetic matrix, in the case of figure 7, is replaced by a paramagnetic, undercritical singlet ground-state matrix, then the effective c_p becomes smaller, but, in contrast to RPA the same is true for the critical ratio predicted in the limit $c \rightarrow 0$.

4. Conclusion

We have developed a theory which simultaneously includes the effects of static inhomogeneities and of thermal disorder. To first order in $1/Z$ the single sites are treated separately and all interact with the same effective medium. The diagrammatic expansion of the τ-ordered two-site Green function is performed in a way which takes direct advantage of this approximate condition.

Our analysis shows that the usual procedure for performing the $1/Z$ expansion to first order adds higher-order contributions to the effective single-site Green function, which are inadequate. This is demonstrated in a direct way by a comparison with the exact value of this quantity obtained in the case of the Ising model. In order to remove these improper contributions the infinite series for $g(\mathrm{i}\omega_n)$ was truncated, so that the inverse Green function, $1/g(\mathrm{i}\omega_n)$, only included the terms to order $1/Z$. The Green functions obtained in this way were found to offer a reasonable account of the phase diagram for $S = 1$ in the non-dilute case. At $c = 1$ the validity of the predictions are probably only limited by the effective-medium approximation itself. As the degree of dilution of the system is increased the accuracy with which the effective single-site Green function is determined becomes more and more important. However, the indications are that the first-order approximation for $g(\mathrm{i}\omega_n)$ is acceptable as long as the effective-medium approximation itself is reasonable, i.e. $c \gtrsim c_\mathrm{p}$. In this regime we also expect the theory to give a fair account of the renormalisation of the energies and the lifetimes of the excitations, in a way which compares in quality with the CPA applied to phonon systems with diagonal disorder.

The spurious divergence of the effective single-site Green function is still present to first order in $1/Z$, but the importance, and thus the inconvenience caused by this divergence at low concentrations, is much reduced in comparison with the zero-order RPA results. An approximation which circumvents this problem is

$$g(\omega) \simeq g_0(\omega)/[1 - \{K(\omega) - K(0)\}g_0(\omega)] \tag{35}$$

which in the non-dilute case coincides with the theory of Lines (1974, 1975) except for his introduction of an effective single-site anisotropy term. This expression neglects thermal contributions to the linewidth, but gives a reasonable estimate of the real part of the self-energy, and at zero frequency $g(0)$ becomes equal to the non-interacting $g_0(0)$, which behaves properly without giving rise to any spurious divergences.

References

Abrikosov A A, Gorkov L P and Dzyaloshinski I E 1963 *Methods of Quantum Field Theory in Statistical Physics* (Englewood Cliffs, NJ: Prentice Hall)
Bak P 1975 *Phys. Rev.* **B12** 5203–12
Care C M and Tucker J W 1977 *J. Phys. C: Solid State Phys.* **10** 2773–82
Elliott R J, Leath B R and Krumhansl J A 1974 *Rev. Mod. Phys.* **46** 465–543
Essam J W 1972 *Phase Transitions and Critical Phenomena* Vol 2 ed. C Domb and M S Green (New York: Academic Press)
Jensen J 1979 *J. Magn. Magn. Mater.* **14** 224–6
—— 1982 *J. Phys. C: Solid State Phys.* **15** 2403–15
Johnson J W and Wang Y-L 1981 *Phys. Rev.* **B24** 5204–13
Lage E J S and Stinchcombe R B 1977 *J. Phys. C: Solid State Phys.* **10** 295–312

Lines M E 1974 *Phys. Rev.* B**9** 3927–31
—— 1975 *Phys. Rev.* B**12** 3766–74
Psaltakis G C and Cottram M G 1982 *J. Phys. C. Solid State Phys.* **15** 4847–68
Schmidt U 1974 *Z. Phys.* **267** 277–82
Stinchcombe R B 1973 *J. Phys. C: Solid State Phys.* **6** 2459–524
Vaks V G, Larkin A I and Pikin S A 1968 *Sov. Phys.-JETP* **26** 647–55
Wang Y-L 1973 *Phys. Rev.* B**8** 2365–6
Whitelaw D J 1981 *J. Phys. C: Solid State Phys.* **14** 2871–86
Yang D H-Y and Wang Y-L 1975 *Phys. Rev.* B**12** 1057–70
Yonezawa F 1968 *Prog. Theor. Phys.* **40** 734–57