Many-body theory of magnetism for ions with complicated level structure

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The problem of incorporating the effects of many-body static spin correlations in excited orbital crystal-field levels is discussed by introducing the concept of a correlated effective field. The resulting theory has the conceptual simplicity of molecular-field theory and can be used for problems where excited orbital crystal-field energies, exchange energies, and thermal energies are all of the same order of magnitude. Correlations are determined by forcing a consistency with the fluctuation theorem and the resulting statistical theory is shown to have an accuracy equivalent to that of random-phase Green's function techniques in the quenched-orbital (spin-only) Heisenberg limit.

I. INTRODUCTION

One of the more difficult statistical problems in many-body magnetism is an adequate description of a lattice of interacting magnetic ions for which thermal energy kT, exchange J, and single-ion crystal-field splittings are all of the same order of magnitude. In particular, we think of those situations for which it is impossible to write a spin Hamiltonian of concise form. These are predominantly systems involving magnetic ions with large unquenched orbital angular momentum. Thus, for example, a system of ferrous or cobaltous ions in quasicubic ligand field and with strong superexchange paths (such as in FeO or CoO) might constitute just such an example at temperatures of the order of (and above) the Néel points.

Such systems, involving a thermal population of excited orbital crystal field levels, are still commonly treated in a molecular-field approximation.[1,2] The reduction of magnetic interactions to effective local fields has the great mathematical advantage of reducing the many-body problem to that of an ensemble of noninteracting effective ions and, in addition, maintains a simple physical picture in terms of single ion energy levels and their perturbation by the local field. In such a local field framework there is no more difficulty in assessing the effect of exchange on excited orbital levels than on the ground orbital levels. The weakness, of course, is the extreme crudeness of the molecular field approximation itself which, by replacing all ions except one by their ensemble averaged states or, for dynamics, small deviations therefrom[2] neglects all interion spin and orbital *static* correlations.

At low temperatures, when it is possible to ignore any thermal population of excited orbital levels, it is always possible to construct a concise spin-Hamiltonian formalism and to pursue spin-wave statistical methods and the like.[3-6] For higher temperatures it is difficult to envisage a tractable formalism which does not in some manner retain the concept of a local field. In this paper we consider the possibility of constructing an equilibrium theory within a local field framework (and which therefore contains much of the conceptual simplicity of molecular field theory itself) which is able to include, in a reasonably sophisticated manner, the important effects of static spin and orbital correlations. To do this we introduce the concept of a "correlated effective field" to take the place of the molecular field. This same concept has been used by the author recently in connection with problems in the theory of lattice dynamics.[7] The present paper is restricted to a discussion of static properties. The corresponding dynamic theory can be generated by linear-response techniques (in analogy with that derived for soft lattice modes in Refs. 7) and differs from time-dependent molecular field theory[2] in the same sense that self-consistent phonon theory[8] differs from time-dependent Hartree theory[9] in lattice dynamics.

Advantage is taken of the fact that conventional molecular field theory violates the fluctuation theorem. We establish below that static correlations can indeed be incorporated into a local-field framework and in such a way that merely requiring the resulting theory to obey a rigorous restriction imposed by the fluctuation theorem determines these correlations completely. We expect the theory to be of greatest value in discussing the effect of correlations on excited orbital levels and in describing disordered but highly correlated motion of magnetic moments with large orbital components.

II. THE CORRELATED EFFECTIVE FIELD

For simplicity we shall consider a system of interacting magnetic ions for which the interion exchange can be written in a simple diagonal bilinear form between *real* spins $\vec{S}$

$$\mathcal{H}_{ex} = -\sum_i \sum_j \sum_\gamma J_{ij}^\gamma S_i^\gamma S_j^\gamma , \tag{2.1}$$

where i and j run over all lattice sites and γ runs over the three orthogonal spatial directions x, y, z. The total magnetic Hamiltonian is assumed to be of the form

$$\mathcal{H}=\sum_i \mathcal{H}_i+\mathcal{H}_{ex}\,, \tag{2.2}$$

where $\mathcal{H}_i$ is the Hamiltonian describing the magnetic ion at site i in the absence of interion exchange J, and includes contributions from intraion Coulomb and exchange forces, crystal field, and spin-orbit origins. In addition, we assume that the eigenvalues E'_{in} and eigenfunctions φ'_{in} of $\mathcal{H}_i$ are known and that the energy separation of the lowest several levels is of order J and kT (at a temperature T of interest).

Consider a particular site i, selected at random. The motion of the spin ($\vec{\mathbf{S}}_i$) and orbital ($\vec{\mathbf{L}}_i$) components of angular momentum at site i can be determined only by those terms in (2.2) which involve i explicitly. It follows that only the equations of motion for spin involve the interactions explicitly. Writing the motion in detail we have

$$\dot{L}_i^x=(i/\hbar)[\mathcal{H}_i, L_i^x]_-\,, \tag{2.3}$$

and

$$\dot{S}_i^x=(i/\hbar)[\mathcal{H}_i, S_i^x]_- - 2\sum_j (J_{ij}^y S_i^z S_j^y - J_{ij}^z S_i^y S_j^z)\,, \tag{2.4}$$

together with the additional equations resulting from a cyclic permutation of x, y, z, (where $[\,,\,]_-$ is a commutator).

In molecular-field theory, the equilibrium equations of motion [Eq. (2.4)], are reduced to single-body form by approximating all $\vec{\mathbf{S}}_j$ by ensemble averages $\langle\vec{\mathbf{S}}_j\rangle$. In contrast, we shall introduce the concept of a *correlated* effective field by replacing each $\vec{\mathbf{S}}_j$ by the sum of two contributions, one its ensemble average, the other a term proportional to the instantaneous deviation of $\vec{\mathbf{S}}_i$ from its own averaged position $\langle\vec{\mathbf{S}}_i\rangle$. Specifically we write (2.4) as

$$\dot{S}_i^x=(i/\hbar)[\mathcal{H}_i, S_i^x]_- - S_i^z h_{corr}^y + S_i^y h_{corr}^z\,, \tag{2.5}$$

where correlated field h_{corr}^γ is assumed to take the form

$$h_{corr}^\gamma = 2\sum_j J_{ij}^\gamma[\langle S_j^\gamma\rangle + A_{ij}^\gamma(S_i^\gamma - \langle S_i^\gamma\rangle)]\,, \tag{2.6}$$

and A_{ij} are temperature-dependent static correlation parameters of as yet unspecified form. Indeed we do not need to know their detailed form since, by writing

$$\sum_j A_{ij}^\gamma J_{ij}^\gamma = \alpha^\gamma \sum_j J_{ij}^\gamma \tag{2.7}$$

to define correlation parameters α^γ ($\gamma=x, y, z$) the effective fields can be expressed in terms of α^γ alone, e.g.,

$$h_{corr}^\gamma = 2\sum_j J_{ij}^\gamma[\langle S_j^\gamma\rangle + \alpha^\gamma(S_i^\gamma - \langle S_i^\gamma\rangle)]\,. \tag{2.8}$$

Since we have not subscripted α^γ, we have taken all lattice sites to be equivalent. In a paramagnetic phase, for which the formalism is simplest and on which we shall concentrate in the present paper, this does not necessarily restrict us to exchange of ferromagnetic sign and, as is physically evident from (2.8), the correlation parameters will tend to be positive for ferromagnets and negative for antiferromagnets.

Consider first the paramagnetic phase in the absence of applied field. Putting ensemble averages equal to zero, using (2.5) and (2.8), and properly symmetrizing to render the observable $\dot{S}_i^x$ Hermitian, we find the "correlated-field" equations of motion

$$\dot{S}_i^x=(i/\hbar)[\mathcal{H}_i, S_i^x]_- - \sum_j (J_{ij}^y\alpha^y - J_{ij}^z\alpha^z)(S_i^y S_i^z + S_i^z S_i^y)\,, \tag{2.9}$$

together with the cyclic permutations. It is now possible to establish an effective (correlated field) Hamiltonian $\mathcal{H}_i^0$(eff) from which the above zero-field equations result in the formal manner

$$\dot{S}_i^\gamma=(i/\hbar)[\mathcal{H}_i^0(\text{eff}), S_i^\gamma]_-\,. \tag{2.10}$$

It is

$$\mathcal{H}_i^0(\text{eff})=\mathcal{H}_i - \sum_j\sum_\lambda J_{ij}^\lambda\alpha^\lambda(S_i^\lambda)^2,\quad \text{zero field}\,, \tag{2.11}$$

where λ runs over the directions x, y, z.

In order to examine magnetic response in the paramagnetic phase, we now introduce an infinitesimal static applied field h_i in direction γ. Taking the susceptibility to be diagonal in the coordinate system x, y, z, we put ensemble averages in directions $\lambda\neq\gamma$ equal to zero. Using (2.5), (2.8), and (2.10), we obtain the correlated field Hamiltonian $\mathcal{H}_i$(eff) in the presence of field h_i in the form

$$\mathcal{H}_i(\text{eff})=\mathcal{H}_i^0(\text{eff}) - h_i\mu_i^\gamma - \sum_j 2J_{ij}^\gamma S_i^\gamma(\langle S_j^\gamma\rangle - \alpha^\gamma\langle S_i^\gamma\rangle)\,, \tag{2.12}$$

with $\mathcal{H}_i^0$(eff) as in (2.11), and where $\vec{\mu}$ signifies magnetic moment.

Thus, with (2.12), we have achieved the reduction of the many-body problem to a single-body (or rather noninteracting ensemble) form. To do so, while maintaining a measure of spin correlations, has cost us in general three temperature-dependent parameters α^x, α^y, α^z, although of course in any particular circumstance, two or even all three may be equal by symmetry. We shall now show, in Secs. III and IV, that even in the most general situation with three different

parameters, they are all determined in an unambiguous fashion by forcing a consistency of the formalism with exact requirements of the fluctuation theorem.

III. MAGNETIC RESPONSE IN THE CORRELATED FIELD APPROXIMATION

In the paramagnetic phase and in the absence of applied field, the correlated effective field Hamiltonian $\mathcal{H}_i^0(\text{eff})$ for the ith magnetic ion is given by (2.11). Its eigenfunctions $\varphi_{in}^0(\alpha)$ and eigenvalues $E_{in}^0(\alpha)$ are readily calculated (as functions of the correlation parameters α^γ) for any particular case. Consider now the application of a static infinitesimal perturbing field parallel to direction γ. The resulting perturbing Hamiltonian $\mathcal{H}_i'$ in the correlated field approximation is, from (2.12),

$$\mathcal{H}_i' = -h_i^\gamma \mu_i^\gamma - \sum_j 2J_{ij}^\gamma S_i^\gamma [\langle S_j^\gamma\rangle - \alpha^\gamma \langle S_i^\gamma\rangle] . \tag{3.1}$$

The resulting perturbed eigenvalues and eigenfunctions to first order of smallness in h_i^γ, $\langle S_j^\gamma\rangle$, $\langle S_i^\gamma\rangle$, are

$$E_{in}(\alpha) = E_{in}^0(\alpha) + (\mathcal{H}_i')_{nn} , \tag{3.2}$$

$$\varphi_{in}(\alpha) = \phi_{in}^0(\alpha) + \sum_{m\neq n} (\mathcal{H}_i')_{mn} \varphi_{im}^0(\alpha) / [E_{in}^0(\alpha) - E_{im}^0(\alpha)] , \tag{3.3}$$

where

$$(\mathcal{H}_i')_{mn} = \langle \varphi_{im}^0(\alpha) | \mathcal{H}_i' | \varphi_{in}^0(\alpha)\rangle , \tag{3.4}$$

and all subsequent subscripted matrix elements also refer to the unperturbed basis $\varphi_n^0(\alpha)$.

The diagonal matrix elements of μ^γ in the perturbed basis are now

$$\langle \varphi_{in}(\alpha) | \mu_i^\gamma | \varphi_{in}(\alpha)\rangle = (\mu_i^\gamma)_{nn} + \sum_{m\neq n} [(\mathcal{H}_i')_{nm}(\mu_i^\gamma)_{mn} + (\mu_i^\gamma)_{nm}(\mathcal{H}_i')_{mn}] / [E_{in}^0(\alpha) - E_{im}^0(\alpha)] . \tag{3.5}$$

Using simple Boltzmann statistics, field-dependent ensemble averages $\langle S_i^\gamma\rangle_h$, $\langle L_i^\gamma\rangle_h$, can now be obtained to first order in small quantities. Putting zero-field averages $\langle S_i^\gamma\rangle_0$, $\langle L_i^\gamma\rangle_0$, and $\langle \mu_i^\gamma\rangle_0$ equal to zero in a paramagnetic phase, we obtain

$$kT\langle S_i^\gamma\rangle_h = h_i \langle \mu_i^\gamma : S_i^\gamma\rangle_0 + \sum_j 2J_{ij}^\gamma [\langle S_j^\gamma\rangle_h - \alpha^\gamma \langle S_i^\gamma\rangle_h] \langle S_i^\gamma : S_i^\gamma\rangle_0 , \tag{3.6}$$

$$kT\langle L_i^\gamma\rangle_h = h_i \langle \mu_i^\gamma : L_i^\gamma\rangle_0 + \sum_j 2J_{ij}^\gamma [\langle S_j^\gamma\rangle_h - \alpha^\gamma \langle S_i^\gamma\rangle_h] \langle S_i^\gamma : L_i^\gamma\rangle_0 , \tag{3.7}$$

where, for arbitrary operators A_i, B_i, we define

$$\langle A_i : B_i\rangle_0 = \sum_n (A_i : B_i)_n \exp[-E_{in}^0(\alpha)/kT] \Big/ \sum_n \exp[-E_{in}^0(\alpha)/kT] , \tag{3.8}$$

in which

$$(A_i : B_i)_n = (A_i)_{nn}(B_i)_{nn} + kT \sum_{m\neq n} [(A_i)_{nm}(B_i)_{mn} + (B_i)_{nm}(A_i)_{mn}] / [E_{im}^0(\alpha) - E_{in}^0(\alpha)] . \tag{3.9}$$

Using the fact that zero-field averages $\langle \cdots\rangle_0$ are site-independent, we now Fourier transform (3.6) and (3.7) with respect to the lattice to obtain

$$kT\langle S^\gamma(\vec{q})\rangle_h = h(\vec{q})\langle \mu_i^\gamma : S_i^\gamma\rangle_0 + 2[J^\gamma(\vec{q}) - \alpha^\gamma J^\gamma(0)]\langle S^\gamma(\vec{q})\rangle_h \langle S_i^\gamma : S_i^\gamma\rangle_0 , \tag{3.10}$$

$$kT\langle L^\gamma(\vec{q})\rangle_h = h(\vec{q})\langle \mu_i^\gamma : L_i^\gamma\rangle_0 + 2[J^\gamma(\vec{q}) - \alpha^\gamma J^\gamma(0)]\langle S^\gamma(\vec{q})\rangle_h \langle S_i^\gamma : L_i^\gamma\rangle_0 , \tag{3.11}$$

in which $L^\gamma(\vec{q})$, $S^\gamma(\vec{q})$, etc. are the Fourier transforms of L_i^γ, S_i^γ, ..., and where

$$J^\gamma(\vec{q}) = \sum_{j-i} J_{ij}^\gamma e^{i\vec{q}\cdot(\vec{i}-\vec{j})} , \tag{3.12}$$

with $\vec{q}$ a wave vector. Equations (3.10) and (3.11) are readily solved for $\langle S^\gamma(\vec{q})\rangle_h$ and $\langle L^\gamma(\vec{q})\rangle_h$ explicitly.

Writing the magnetic-moment vector (in units of Bohr magneton)

$$\vec{\mu} = 2\vec{S} + k\vec{L} \tag{3.13}$$

in terms of the real spin and orbital angular-momentum vectors (allowing for an orbital reduction k of scalar form), the wave-vector-dependent susceptibility $\chi^\gamma(\vec{q})$ now follows directly as

$$\chi^\gamma(\vec{q}) = \langle 2S^\gamma(\vec{q}) + kL^\gamma(\vec{q})\rangle_h / h(\vec{q}) . \tag{3.14}$$

After a little algebra we find

$$kT\chi^\gamma(\vec{q}) = \langle \mu_i^\gamma : \mu_i^\gamma\rangle_0 + U^\gamma(\vec{q}) , \tag{3.15}$$

where

$$U^\gamma(\vec{q}) = \frac{2[J^\gamma(\vec{q}) - \alpha^\gamma J^\gamma(0)]\langle \mu_i^\gamma : S_i^\gamma\rangle^2}{kT - 2[J^\gamma(\vec{q}) - \alpha^\gamma J^\gamma(0)]\langle S_i^\gamma : S_i^\gamma\rangle_0} . \tag{3.16}$$

We shall now establish, via the fluctuation theorem that $\sum_{\vec{q}} U^\gamma(\vec{q}) = 0$. Using (3.16), this immediately determines the correlation parameters as

$$\alpha^\gamma = \sum_{\vec{q}} \eta^\gamma(\vec{q}) J^\gamma(\vec{q}) \Big/ \sum_{\vec{q}} \eta^\gamma(\vec{q}) J^\gamma(0) , \tag{3.17}$$

where

$$[\eta^\gamma(\vec{q})]^{-1} = kT - 2[J^\gamma(\vec{q}) - \alpha^\gamma J^\gamma(0)]\langle S_i^\gamma : S_i^\gamma\rangle_0 . \tag{3.18}$$

The equations (3.18), (3.17), and (2.11), the last of which is required to generate the zero-field-correlated single-ion eigenvalues and eigenfunctions necessary to evaluate $\langle S_i^\gamma : S_i^\gamma\rangle_0$, now close the problem and allow for the determination of static paramagnetic properties in the correlated effective-field approximation without more ado.

The general formalism set out above takes on a particularly simple form for high-symmetry situations. An example might be for CoO in its (cubic) paramagnetic phase. We note from (2.11) that for a system which has, by symmetry, $J_{ij}^x = J_{ij}^y = J_{ij}^z = J_{ij}$ and $\alpha^x = \alpha^y = \alpha^z = \alpha$, the correlated effective-field Hamiltonian $\mathcal{H}_i^0(\text{eff})$ differs from $\mathcal{H}_i$ by only a constant. This means that local ith ion eigenfunctions and energy separations are not functions of α and that the entire self-consistency problem reduces to a single implicit equation (3.17) for α.

We now establish the required relationship $\sum_{\vec{q}} U(\vec{q}) = 0$ by a discussion of the fluctuation theorem in the present context.

IV. THE FLUCTUATION THEOREM

Consider the exact many-body Hamilton $\mathcal{H}$ in the presence of an applied field in the direction γ. It can be written

$$\mathcal{H} = \mathcal{H}_0 - \sum_i \mu_i^\gamma h_i^\gamma , \tag{4.1}$$

where $\mathcal{H}_0$ is the zero-field Hamiltonian. Introducing the wave vector $\vec{q}$, we can rewrite (4.1) as

$$\mathcal{H} = \mathcal{H}_0 - \sum_{\vec{q}} \mu^\gamma(-\vec{q}) h^\gamma(\vec{q}) . \tag{4.2}$$

By definition, the ensemble average $\langle \mu^\gamma(\vec{q})\rangle_h$ is

$$\langle \mu^\gamma(\vec{q})\rangle_h = \mathrm{Tr}[\mu^\gamma(\vec{q}) e^{-\beta\mathcal{H}}]/\mathrm{Tr}(e^{-\beta\mathcal{H}}) , \tag{4.3}$$

where

$$\beta = 1/kT . \tag{4.4}$$

The wave-vector-dependent susceptibility $\chi^\gamma(\vec{q})$ now follows from (4.3) by direct differentiation with respect to $h^\gamma(\vec{q})$. Recognizing that $\sum_{\vec{q}} \mu^\gamma(-\vec{q}) h^\gamma(\vec{q})$ does not in general commute with the zero-field Hamiltonian, we proceed as follows.

By direct differentiation we verify that

$$\frac{\partial}{\partial\beta} e^{\beta\mathcal{H}_0} e^{-\beta\mathcal{H}} = e^{\beta\mathcal{H}_0} \sum_{\vec{q}} \mu^\gamma(-\vec{q}) h^\gamma(\vec{q}) e^{-\beta\mathcal{H}} . \tag{4.5}$$

It follows, to first order in applied field, that

$$e^{-\beta\mathcal{H}} = e^{-\beta\mathcal{H}_0} \times\left(1 + \int_0^\beta e^{\theta\mathcal{H}_0} \sum_{\vec{q}} \mu^\gamma(-\vec{q}) h^\gamma(\vec{q}) e^{-\theta\mathcal{H}_0} d\theta\right) , \tag{4.6}$$

and therefore

$$\frac{\partial}{\partial h^\gamma(\vec{q})} e^{-\beta\mathcal{H}} = e^{-\beta\mathcal{H}_0} \int_0^\beta e^{\theta\mathcal{H}_0} \mu^\gamma(-\vec{q}) e^{-\theta\mathcal{H}_0} d\theta , \tag{4.7}$$

to lowest order. Using (4.7) one can differentiate (4.3) with respect to $h^\gamma(\vec{q})$ in straightforward fashion to find

$$\chi^\gamma(\vec{q}) = \langle \int_0^\beta e^{\theta\mathcal{H}_0} \mu^\gamma(-\vec{q}) e^{-\theta\mathcal{H}_0} \mu^\gamma(\vec{q}) d\theta\rangle_0 - \beta\langle \mu^\gamma(-\vec{q})\rangle_0 \langle \mu^\gamma(\vec{q})\rangle_0 , \tag{4.8}$$

where $\langle\cdots\rangle_0$ indicates an ensemble average with respect to the zero-field Hamiltonian, e.g.,

$$\langle\cdots\rangle_0 = \mathrm{tr}[(\cdots) e^{-\beta\mathcal{H}_0}]/\mathrm{tr}[e^{-\beta\mathcal{H}_0}] . \tag{4.9}$$

In a paramagnetic phase $\langle \mu^\gamma(\vec{q})\rangle = 0$ by definition and (4.8) reduces to

$$\chi^\gamma(\vec{q}) = \langle \int_0^\beta e^{\theta\mathcal{H}_0} \mu^\gamma(-\vec{q}) e^{-\theta\mathcal{H}_0} \mu^\gamma(\vec{q}) d\theta\rangle_0 , \tag{4.10}$$

a result exact to lowest order in field. Summing over all allowed $\vec{q}$ values in the first Brillouin zone of the reciprocal lattice, we obtain

$$\sum_{\vec{q}} \chi^\gamma(\vec{q}) = N\langle \int_0^\beta e^{\theta\mathcal{H}_0} \mu_i^\gamma e^{-\theta\mathcal{H}_0} \mu_i^\gamma d\theta\rangle_0 , \tag{4.11}$$

where there are N magnetic ions in the macroscopic lattice. In the correlated field approximation, we now recognize the equivalence of $\mathcal{H}_0$ and $\mathcal{H}_i^0(\text{eff})$ of (2.11) for calculating ensemble averages involving a single (ith) magnetic ion alone. In particular, we can approximate

$$\langle \int_0^\beta e^{\theta\mathcal{H}_0} \mu_i^\gamma e^{-\theta\mathcal{H}_0} \mu_i^\gamma d\theta\rangle_0 = \langle \int_0^\beta e^{\theta\mathcal{H}_i^0(\text{eff})} \mu_i^\gamma e^{-\theta\mathcal{H}_i^0(\text{eff})} \mu_i^\gamma d\theta\rangle_0 . \tag{4.12}$$

Using the basis $\varphi_{in}^0(\alpha)$ for which the Hamiltonian $\mathcal{H}_i^0(\text{eff})$ is diagonal, and hence $\exp[\theta\mathcal{H}_i^0(\text{eff})] = \exp[\theta E_{in}^0(\alpha)]$ is also diagonal, the right-hand side of Eq. (4.12) can be expanded and integrated directly to obtain

$$\langle \int_0^\beta e^{\theta\mathcal{H}_i^0(\text{eff})} \mu_i^\gamma e^{-\theta\mathcal{H}_i^0(\text{eff})} \mu_i^\gamma d\theta\rangle_0 = \beta\langle \mu_i^\gamma : \mu_i^\gamma\rangle_0 , \tag{4.13}$$

where the colon product symbolism has been defined earlier in (3.8) and (3.9). Using (4.11)–(4.13) we obtain finally the required fluctuation result

$$\sum_{\vec{q}} kT\chi^\gamma(\vec{q}) = N\langle \mu_i^\gamma : \mu_i^\gamma\rangle_0 , \tag{4.14}$$

which, together with (3.15), provides us with the relationship

$$\sum_{\vec{q}} U^\gamma(\vec{q}) = 0 , \tag{4.15}$$

which was used in Sec. III to close the correlated effective field equations.

V. THE QUENCHED LIMIT AND A CONTACT WITH GREEN'S-FUNCTION THEORY

In order to obtain some idea of the accuracy of the correlated effective field approximation and to put the present method into some kind of many-body perspective, it is useful to use it to solve a ferromagnetic problem which is already susceptible to more conventional techniques. Consider, for example, the simple case of an isotropic Heisenberg system with fully quenched orbital angular momentum. The relevant Hamiltonian is

$$\mathcal{H} = -\sum_i \sum_j J_{ij} \vec{S}_i \cdot \vec{S}_j \ , \tag{5.1}$$

with local ion Hamiltonian $\mathcal{H}_i$ of (2.2) replaced by a constant, and with local magnetic moment $\vec{\mu}_i = 2\vec{S}_i$, $\vec{L}_i = 0$.

It follows immediately from symmetry that $\alpha^x = \alpha^y = \alpha^z = \alpha$ which, in turn [using (2.11)] leads to the finding $\mathcal{H}_i^0(\text{eff}) = \text{constant}$. One can therefore choose a representation for S_i^γ which is diagonal and, using (3.8), obtain

$$\langle S_i^\gamma : S_i^\gamma \rangle_0 = \sum_{-S}^{+S} M^2 = S(S+1)/3 \ . \tag{5.2}$$

Putting $L_i^\gamma = 0$ and using (3.15) and (3.16), we find the expression for wave-vector-dependent susceptibility $\chi(\vec{q})$ (where superscript γ can now be dropped in view of the equivalence of directions x, y, z) in the form

$$\tfrac{4}{3} S(S+1)[\chi(\vec{q})]^{-1} = kT - \tfrac{2}{3} S(S+1)[J(q) - \alpha J(0)] \ . \tag{5.3}$$

It follows that the static uniform susceptibility is given by

$$\tfrac{4}{3} S(S+1)[\chi(0)]^{-1} = kT - \tfrac{2}{3} S(S+1) J(0)(1-\alpha) \ , \tag{5.4}$$

and that correlation parameter α can be eliminated between (5.3) and (5.4) to obtain

$$[\chi(\vec{q})]^{-1} - [\chi(0)]^{-1} = \tfrac{1}{2}[J(0) - J(\vec{q})] \ . \tag{5.5}$$

From (3.15) and (4.15) we have

$$N^{-1} \sum_{\vec{q}} \chi(\vec{q}) = 4S(S+1)/3kT \ , \tag{5.6}$$

which finally, using (5.5), leads to an equation relating uniform static susceptibility directly to temperature, namely

$$N^{-1} \sum_{\vec{q}} \{4[\chi(0)]^{-1} + 2[J(0) - J(\vec{q})]\}^{-1} = S(S+1)/3kT \ . \tag{5.7}$$

In particular, for the ferromagnetic situation with positive J, the uniform susceptibility diverges at the Curie temperature T_c given by

$$N^{-1} \sum_{\vec{q}} [J(0) - J(\vec{q})]^{-1} = 2S(S+1)/3kT_C \ . \tag{5.8}$$

Both (5.7) and (5.8) are results familiar from the random-phase Green's-function approximation[10,11] for this simple spin-only magnetic situation. We conclude that the correlated effective field approximation is equivalent to random-phase-approximation Green's-function theory in the (paramagnetic) quenched-orbital Heisenberg limit. More generally a closer correspondence is with the spherical model, an increased accuracy over the RPA Green's-function theory being evident for anisotropic exchange and particularly marked for the Ising limit.

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