[7]J. von Heimburg and H. Thomas, to be published.

[8]Note that less divergent contributions which are of the type

$$b^{2l}\sum_{n=1}^{n_0} K^{-n}\gamma^{(2l-r)n}$$

give rise to terms like $|b|^{K+r}$.

[9]There is no apparent way to obtain a closed expression for $A(t)$ in Eq. (8).

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Singlet-Singlet Induced-Moment System*

D. A. Pink†
Department of Physics, University of Guelph, Guelph, Ontario, N1G 2W1, Canada
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A new decoupling procedure for the two-level induced-moment system in the paramagnetic regime is described. It offers improvements over the results of other methods and can in principle be extended to other systems.

It is becoming commonplace to study and understand the magnetic properties of rare-earth metals and intermetallic compounds in which the effects of many crystal-field states are significant. These systems are studied theoretically by using a molecular-field approximation,[1] a random-phase approximation (RPA),[2] or, less frequently, the two-site correlation approximation (TSCA).[3] One of the original systems studied was the two-singlet induced-moment system[1-3] in which the two lowest single-ion eigenstates, $|A\rangle$ and $|B\rangle$, of the crystal-field operator V^c are magnetic singlets separated by an energy Δ. All matrix elements of the total magnetic moment operator $\vec{J}_i$ of the ith ion between these two states are zero except for $\langle B|J^z|A\rangle=\alpha$. The Hamiltonian was assumed to be

$$H=-\textstyle\sum_{ij}K_{ij}\vec{J}_i\cdot\vec{J}_j+\sum_j V_j^c, \quad V^c|A\rangle=0, \quad V^c|B\rangle=\Delta|B\rangle. \tag{1}$$

The collective excitations have been calculated using the RPA and TSCA, and some thermodynamic quantities have been calculated self-consistently. Two points should be noted, one of which will be taken up: (a) The magnetization is double valued as a function of temperature T, in general, which is interpreted as indicating a first-order phase transition. There are arguments that the transition should be second order. (b) The exact excitation spectrum in one dimension has been calculated.[4] It does not agree with RPA or TSCA calculations. Here, a decoupling scheme will be outlined which gives results for the paramagnetic region[4] ($\langle J^z\rangle=0$) which agree closely with the exact result. The extension of it to describe the magnetically ordered region will be treated elsewhere.

For simplicity we shall assume a nearest-neighbor exchange interaction and map the operators $J_j^{\pm}$, J_j^z, and V_j^c onto the components of a pseudospin-$\frac{1}{2}$ operator, $\vec{\sigma}_j$. The eigenstates of σ_j^z are $|\pm\rangle_j$ belonging to eigenvalues $\pm\frac{1}{2}$; we map $|A\rangle\rightarrow|+\rangle$ and $|B\rangle\rightarrow|-\rangle$, and the Hamiltonian becomes[2,3,5]

$$H=-J\textstyle\sum_{j\delta}\sigma_j^x\sigma_{j+\delta}^x-\Delta\sum_j\sigma_j^z+\frac{1}{2}\Delta N, \tag{2}$$

where $\vec{\delta}$ is a primitive lattice vector and N is the total number of ions. The operators σ_j^x and σ_j^y are two of the single-excitation operators of this system. In the paramagnetic region $\langle\sigma^x\rangle=0$. The equations of motion for the Green's functions are[6]

$$\begin{aligned}
&E\langle\langle\sigma_i^x|B_j\rangle\rangle=g^x(\vec{r}_j-\vec{r}_i)+i\Delta\langle\langle\sigma_i^y|B_j\rangle\rangle,\\
&E\langle\langle\sigma_i^y|B_j\rangle\rangle=g^y(\vec{r}_j-\vec{r}_i)-i\Delta\langle\langle\sigma_i^x|B_j\rangle\rangle+2iJ\textstyle\sum_\delta\langle\langle\sigma_i^z\sigma_{i+\delta}^x|B_j\rangle\rangle,\\
&g^\nu(\vec{r}_j-\vec{r}_i)=(2\pi)^{-1}\langle[\sigma_i^\nu,B_j]\rangle.
\end{aligned} \tag{3}$$

At this point the RPA replaces $\langle\langle\sigma_i^z\sigma_{i+\delta}^x|B_j\rangle\rangle$ by $\langle\sigma^z\rangle\langle\langle\sigma_{i+\delta}^x|B_j\rangle\rangle$. This approach treats σ_i^z as an approximate constant of the motion by taking the approximate ground state of (2) to be $\prod_j|+\rangle_j$. The ground state for the case $N=2$ contains $|+\rangle_1|+\rangle_2$ with a (J/Δ)-dependent admixture of $|-\rangle_1|-\rangle_2$. If we consider

the equations for the Green's functions of this two-site system and ask how we can decouple them in order to obtain only the single-excitation energies, we see that we can decouple as follows:

$$\langle\langle \sigma_i^z \sigma_{i+\delta}^x | B_j \rangle\rangle \equiv \langle\langle [U_i - (J/\Delta)\sigma_i^x \sigma_{i+\delta}^x]\sigma_{i+\delta}^x | B_j \rangle\rangle \rightarrow \langle U_i \rangle \langle\langle \sigma_{i+\delta}^x | B_j \rangle\rangle - (J/4\Delta)\langle\langle \sigma_i^x | B_j \rangle\rangle, \tag{4}$$

$$U_i \equiv \sigma_i^z + (J/\Delta)\sigma_i^x \sigma_{i+\delta}^x, \quad i, i+\delta = 1, 2,$$

where the U_i have been decoupled because the ground state is an eigenstate of them. If we evaluate $\langle U_i \rangle$ in the ground state we obtain the energies between the ground and first and second excited states. Accordingly, we shall use the following decoupling scheme in (3):

$$\langle\langle \sigma_i^z \sigma_{i+\delta}^x | B_j \rangle\rangle \rightarrow U \langle\langle \sigma_{i+\delta}^x | B_j \rangle\rangle - (J/4\Delta)\langle\langle \sigma_i^x | B_j \rangle\rangle,$$

$$U = \langle U_i \rangle, \quad U_i \equiv \sigma_i^z + (J/\Delta)\sigma_i^x \sigma_{i+\delta}^x, \tag{5}$$

where the U_i is taken as an approximate constant of motion. By Fourier transforming with respect to wave vector $\vec{\lambda}$ and choosing B_j appropriately we obtain

$$\langle \sigma^z \rangle [(1+C)I_0 - DI_1] = 1, \quad \langle \sigma_i^x \sigma_{i+\delta}^x \rangle = \tfrac{1}{2}\langle \sigma^z \rangle I_1, \quad \langle (\sigma^-)^2 \rangle = \tfrac{1}{2}\langle \sigma^z \rangle [DI_1 - (C-1)I_0], \quad C = 1 + J^2 z/2\Delta^2,$$

$$D = (2Jz/\Delta)U, \quad I_n = (\Delta/N)\textstyle\sum_{\vec{\lambda}} [\gamma_{\vec{\lambda}}^n / E(\vec{\lambda})] \coth[\tfrac{1}{2}\beta E(\vec{\lambda})], \quad \gamma_{\vec{\lambda}} = z^{-1} \textstyle\sum_{\vec{\delta}} \exp(i\vec{\lambda}\cdot\vec{\delta}), \quad E^2(\vec{\lambda}) = \Delta^2(C - D\gamma_{\vec{\lambda}}), \tag{6}$$

where z is the number of nearest neighbors. There are two points to note: (i) The wave-vector–independent part of the spectrum is $\Delta^2 + J^2 z/2$ and not Δ^2 (RPA) or $\Delta^2 + 8J\Delta\langle \sigma^z \rangle \langle \sigma_i^x \sigma_{i+\delta}^x \rangle$ (TSCA). This result is identical to the exact result in one dimension. No other approximation, apparently, gives this. (ii) The wave-vector–dependent part of $E(\vec{\lambda})$ contains U, where the RPA and TSCA have $\langle \sigma^z \rangle$ and the exact result is $\frac{1}{2}$. In one dimension when $T = 0$ some calculations give

J/Δ	$\langle \sigma^z \rangle$	$\langle \sigma_i^x \sigma_{i+\delta}^x \rangle$	$\langle (\sigma^-)^2 \rangle$	U
0.8	0.404	0.124	0.029	0.503
0.7	0.433	0.099	0.013	0.502
0.6	0.453	0.081	0.005	0.501
0.45	0.474	0.058	0.001	0.500
0.45 (RPA)	0.460		0.065	

and the critical values of Δ/J at which the energy gap goes to zero are 2.0 (molecular field), 1.92 (RPA), 1.0 (exact), and 1.1 (here). Attention is drawn to the small values of $\langle (\sigma^-)^2 \rangle$, even near the critical value of $J/\Delta \simeq 0.91$, and the closeness of U to $\frac{1}{2}$.

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†Permanent address: Department of Physics, St. Francis Xavier University, Antigonish, Nova Scotia, Canada.

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