Second-order Green's-function approach for the study of the sensitivity of the Curie temperature to single-ion anisotropy

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This paper utilizes the second-order Green's-function formalism to treat a Heisenberg ferromagnet in the presence of single-ion crystal-field anisotropy represented by the Hamiltonian: $H = -J \sum_{i,\delta} (\vec{S}_i \cdot \vec{S}_{i+\delta}) - D \sum_i (S_i^z)^2$. Second-order equations of motion for the Green's functions $\langle\langle S_i^z; S_j^z \rangle\rangle$ and $\langle\langle S_i^+; S_j^- \rangle\rangle$ are developed and the higher-order Green's functions are decoupled in the zeroth-order approximation in which the interspin correlations are not taken into account rigorously. The spin-correlation functions are derived and are solved self-consistently in the limit of zero spontaneous magnetization. The Curie temperature is thus obtained. Calculations are restricted to a simple-cubic lattice and positive values of D only, and the sensitivity of the Curie temperature to the single-ion anisotropy is critically examined for a spin-1 lattice. It is seen that the results agree very closely with those of the Green's-function diagram technique. The results are also compared with those of the first-order Green's-function theory using the random-phase approximations of Lines using the correlated-effective-field approximation, and of the molecular-field theory.

I. INTRODUCTION

Magnetic systems with single-ion crystal-field anisotropy are of current theoretical interest due to its presence in many ferro- and antiferromagnetic compounds, especially in the rare-earth compounds and in the complex salts of transition-metal elements.[1] The problem has been treated by mean-field approximation,[2] spin-wave approximation,[3] high-temperature series expansion,[4] cluster-approximation[5] Green's-function equation-of-motion method,[6] standard basis-operator techniques,[7] and latest by the Green's-function diagram method.[8] Of all the above mentioned approximational methods, the results of the Green's-function diagrammatic technique[8] are found to be most accurate. The present paper contains an attempt to understand the statistical mechanics of a uniaxial ferromagnet using second-order Green's-function theory.[9-14]

It has been found that the second-order Green's-function theory is a better tool in dealing with the paramagnetic phase of various magnetic systems and the results obtained by the theory for the paramagnetic specific heat and paramagnetic susceptibility are, indeed, better[12] than those obtained from a first-order theory within the same decoupling framework. It may, however, be noted that when one tries to apply the second-order theory to deal with the spontaneous magnetization phase, the procedure becomes severely complicated owing to the appearance of the

magnetization parameter $\langle S^z \rangle$. To our knowledge, no such calculation has been carried out even for a simple case like an isotropic Heisenberg ferromagnet. Although it would have been interesting to study some typical aspects of the low-temperature phase, such as the variation of m with respect to the single-ion anisotropy, we, at present, have restricted our calculations to a relatively simpler problem, namely, the variation of the Curie temperature T_C with respect to the single-ion anisotropy. The present method involves the derivation of the expressions for the correlation functions, which, in the limit $m \to 0$, yield the Curie temperature. Computations are carried out for a simple-cubic lattice and all values of spin and the results for a spin-1 lattice are compared with those of previous authors.

II. SECOND-ORDER GREEN'S FUNCTIONS

A uniaxial Heisenberg ferromagnet with N localized spins is represented by the Hamiltonian

$$H = -J \sum_{i,\delta} \vec{S}_i \cdot \vec{S}_{i+\delta} - D \sum_i (S_i^z)^2 \ , \qquad (1)$$

where J is the nearest-neighbor exchange constant, D is the single-ion anisotropy parameter, and $\vec{\delta}$ is a nearest-neighbor vector. We have restricted our calculations for positive values of D only.

The first-order equation of motion for the Green's

function $\langle\langle S_i^z; S_j^z\rangle\rangle$ is

$$E\langle\langle S_i^z; S_j^z\rangle\rangle = J\sum_\delta \langle\langle (S_{i+\delta}^+ S_i^- - S_i^+ S_{i+\delta}^-); S_j^z\rangle\rangle \quad . \tag{2}$$

One can develop the second-order equation by writing down similar equations of motion for the Green's functions occurring in the right-hand side of Eq. (2). The result is

$$E^2\langle\langle S_i^z; S_j^z\rangle\rangle = \frac{J}{\pi}\sum_\delta \{\langle S_i^+ S_{i+\delta}^-\rangle(\delta_{ij} - \delta_{i+\delta,j})\}$$

$$+ 2J^2 \sum_{\delta,\delta'} \langle\langle [(B_{i+\delta',i+\delta} + B_{i+\delta,i+\delta'})S_i^z - (B_{i+\delta+\delta',i} + B_{i,i+\delta+\delta'})S_{i+\delta}^z - (B_{i+\delta',i} + B_{i,i+\delta'})A_{i+\delta,\delta'}]; S_j^z\rangle\rangle$$

$$+ DJ \sum_\delta \langle\langle (S_i^+ L_{i+\delta}^- + S_i^- L_{i+\delta}^+ - S_{i+\delta}^+ L_i^- - S_{i+\delta}^- L_i^+); S_j^z\rangle\rangle \quad , \tag{3}$$

where we have used the notations

$$B_{i,j} = S_i^+ S_j^- , \quad A_{i+\delta,\delta'} = S_{i+\delta}^z - S_{i+\delta+\delta'}^z \quad ,$$

and

$$L_i^\pm = S_i^z S_i^\pm + S_i^\pm S_i^z \quad .$$

From the above equation of motion we observe that two kinds of higher-order Green's functions are to be decoupled. These are $\langle\langle B_{f,g} S_i^z; S_m^z\rangle\rangle$ and $\langle\langle S_f^\pm L_g^\mp; S_m^z\rangle\rangle$. We employ some random-phase approximation (RPA)-like approximations for decoupling these Green's functions. In the first kind, a factor $\langle B_{f,g}\rangle$ simply separates out, so that

$$\langle\langle B_{f,g} S_i^z; S_m^z\rangle\rangle = \langle B_{f,g}\rangle \langle\langle S_i^z; S_m^z\rangle\rangle \quad . \tag{4}$$

To decouple other types of Green's functions, we note that the quantity $S^z S^+ + S^+ S^z$ goes as $(2S-1)S^+$ at low temperatures, so that one can assume that the quantity $S^z S^+ + S^+ S^z$ at any higher temperature goes as $\Phi(T)(2S-1)S^+$, where $\Phi(T)$ is a temperature-renormalization function. So, we can decouple the Green's functions of the second kind in the manner

$$\langle\langle S_f^-(S_g^+ S_g^z + S_g^z S_g^+); S_j^z\rangle\rangle = 2\Phi(T)(2S-1)\langle\langle S_g^z; S_j^z\rangle\rangle \quad . \tag{5a}$$

In a similar first-order theory, $\Phi(T)$ is usually chosen to be $\langle S^z\rangle/S$ where spin correlations are not taken

into account rigorously.[15] From this analogy we shall assume $\Phi(T)$ to be equal to $\langle S_f^- S_g^+\rangle/S$ for the present case and the decoupling finally reads

$$\langle\langle S_f^-(S_g^+ S_g^z + S_g^z S_g^+); S_j^z\rangle\rangle = 2Q\langle S_f^- S_g^+\rangle \langle\langle S_g^z; S_j^z\rangle\rangle \quad , \tag{5b}$$

where $Q = (2S-1)/S$.

Utilizing the above decoupling procedures and then making usual Fourier transforms, one obtains

$$G_k^z(E) = (J/\pi) z S_1 (1 - \gamma_k)/[E^2 - E_k^2(1)] \quad , \tag{6}$$

where $E_k(1)$ represents one kind of the energy spectrum given by

$$E_k^2(1) = 4J^2 z^2 S_1(1 - \gamma_k)[(1/\eta) - \gamma_k] \quad , \tag{7}$$

$$1/\eta = (1/\mu^1) - DQ/Jz \quad , \tag{8}$$

$$1/\mu^1 = (zS_1)^{-1} \sum_\delta \langle S_0^- S_{\delta+\delta'}^+\rangle \quad , \tag{9}$$

$$\gamma_k = z^{-1} \sum_\delta \exp(i\vec{k}\cdot\vec{\delta}) \quad , \tag{10}$$

and $S_1 = \langle S_0^- S_\delta^+\rangle$. We now define the Green's function

$$G_{ij}(E) = \langle\langle S_i^+; S_j^-\rangle\rangle \tag{11}$$

and for this Green's function one gets the following first-order equation:

$$E\langle\langle S_i^+; S_j^-\rangle\rangle = \frac{1}{\pi}\langle S^z\rangle\delta_{ij} + 2J\sum_\delta \langle\langle (S_{i+\delta}^z S_i^+ - S_i^z S_{i+\delta}^+); S_j^-\rangle\rangle + D\langle\langle (S_i^z S_i^+ + S_i^+ S_i^z); S_j^-\rangle\rangle \quad . \tag{12}$$

Writing the equations of motion for the Green's functions

$$\langle\langle S_{i+\delta}^z S_i^+; S_j^-\rangle\rangle, \quad \langle\langle S_i^z S_{i+\delta}^+; S_j^-\rangle\rangle, \quad \langle\langle (S_i^z S_i^+ + S_i^+ S_i^z); S_j^-\rangle\rangle \quad ,$$

and substituting them in Eq. (12) we get

$$E^2 \langle\langle S_i^+; S_j^- \rangle\rangle = \frac{E}{\pi} \langle S^z \rangle \delta_{ij} + \frac{D}{\pi}(2C_0 - S_0)\delta_{ij} + \frac{J}{\pi} \sum_\delta [(2C_1 + S_1)(\delta_{ij} - \delta_{i+\delta,j})]$$

$$+ 2J^2 \sum_{\delta,\delta'} \langle\langle [2(S_{i+\delta}^z L_{i+\Delta}^z - S_i^z L_{i+\delta+\delta'}^z) + (B_{i+\delta+\delta',i+\delta'} - B_{i+\delta,i+\delta+\delta'})S_i^+ + (B_{i,i+\delta} - B_{i+\delta,i})S_{i+\delta'}^+]; S_j^- \rangle\rangle$$

$$+ 2JD \sum_\delta \langle\langle [S_{i+\delta}^z L_i^+ - S_i^z L_{i+\delta}^+ - B_{i,i+\delta}S_i^+ - 2S_i^+ S_i^z S_i^z + S_{i+\delta}^z L_i^+ + \tfrac{1}{2}S_{i+\delta}^+ (S_i^+ S_i^- + S_i^- S_i^+)]; S_j^- \rangle\rangle$$

$$+ D^2 \langle\langle (S_i^z L_i^+ + L_i^+ S_i^z); S_j^- \rangle\rangle \ , \tag{13}$$

where

$$C_0 = \langle S_0^z S_0^z \rangle, \quad S_0 = \langle S_0^- S_0^+ \rangle, \quad C_1 = \langle S_0^z S_\delta^z \rangle, \quad L_{i+\Delta}^z = S_{i+\delta}^z S_i^+ - S_i^z S_{i+\delta}^+ \ ,$$

and

$$L_{i+\delta+\delta'}^z = S_{i+\delta+\delta'}^z S_{i+\delta'}^+ - S_{i+\delta'}^z S_{i+\delta+\delta'}^+ \ .$$

The Green's functions $\langle\langle S_f^z L_g^+; S_m^- \rangle\rangle$ and $\langle\langle L_f^+ S_g^z; S_m^- \rangle\rangle$ can be decoupled in the manner analogous to Eq. (5). The decoupling now reads as

$$\langle\langle S_f^z L_g^+; S_m^- \rangle\rangle = 2Q \langle S_f^z S_g^z \rangle \langle\langle S_g^+; S_m^- \rangle\rangle \ , \tag{14}$$

$$\langle\langle L_f^+ S_g^z; S_m^- \rangle\rangle = 2Q \langle S_f^z S_g^z \rangle \langle\langle S_f^+; S_m^- \rangle\rangle \ . \tag{15}$$

To decouple the other Green's functions, we shall adopt the following RPA schemes:

$$\langle\langle S_f^z S_g^z S_l^+; S_m^- \rangle\rangle = \langle S_f^z S_g^z \rangle \langle\langle S_l^+; S_m^- \rangle\rangle \ , \tag{16}$$

$$\langle\langle S_f^+ (S_g^+ S_g^- + S_g^- S_g^+); S_m^- \rangle\rangle = 2 \langle S_f^+ S_g^- \rangle \langle\langle S_g^+; S_m^- \rangle\rangle + (\langle S_g^+ S_g^- \rangle + \langle S_g^- S_g^+ \rangle) \langle\langle S_f^+; S_m^- \rangle\rangle \ , \tag{17}$$

$$\langle\langle S_f^+ S_g^+ S_l^-; S_m^- \rangle\rangle = \langle S_f^+ S_l^- \rangle \langle\langle S_g^+; S_m^- \rangle\rangle + \langle S_g^+ S_l^- \rangle \langle\langle S_f^+; S_m^- \rangle\rangle \ . \tag{18}$$

Utilizing these decoupling approximations and making the usual Fourier transforms, one obtains

$$G_k(E) = [(m/\pi)E + (2/\pi)JzC_{11}(d - \gamma_k)]/[E^2 - E_k^2(2)] \ , \tag{19}$$

where $E_k(2)$ represents the second kind of energy spectrum

$$E_k^2(2) = 4J^2 z^2 C_{11}(\alpha - \beta'\gamma_k + \gamma_k^2) \ , \tag{20}$$

$$\alpha = \frac{1}{\mu} + \frac{DQ}{JzC_{11}} \left[2C_1 - \frac{S_1}{2Q} + \frac{DC_0}{Jz} \right] \ , \tag{21}$$

$$\beta' = 1 + \frac{1}{\mu} + \frac{DQ}{JzC_{11}}[C_1 + (C_0 - \tfrac{1}{2}S_0)/Q] \ , \tag{22}$$

$$d = 1 + D(C_0 - \tfrac{1}{2}S_0)/(JzC_{11}) \ , \tag{23}$$

$$1/\mu = (zC_{11})^{-1} \sum_\delta (\langle S_0^z S_{\delta+\delta'}^z \rangle + \tfrac{1}{2} \langle S_0^- S_{\delta+\delta'}^+ \rangle) \ , \tag{24}$$

$$m = \langle S^z \rangle \ ,$$

and

$$C_{11} = C_1 + S_1/2 \ .$$

Equations (21) and (22) show that for small D/J we can reasonably assume

$$\beta' = 1 + \alpha \tag{25}$$

to simplify the above complicated equations.

III. CURIE TEMPERATURE

Substituting the Green's functions of Eqs. (6) and (19) in the spectral theorem

$$\langle BA \rangle = \lim_{\epsilon \to 0} i \int_{-\infty}^\infty (\langle\langle A; B \rangle\rangle_{E+i\epsilon} - \langle\langle A; B \rangle\rangle_{E-i\epsilon})(e^{\beta E} - 1)^{-1} dE \ , \tag{26}$$

one obtains the following expressions for the parallel and perpendicular spin-correlation function:

$$\langle S_0^z S_g^z \rangle = JzS_1 \frac{1}{N} \sum_k \frac{1-\gamma_k}{E_k(1)} \coth\left[\tfrac{1}{2}\beta E_k(1)\right] \exp(-i\vec{k}\cdot\vec{g}) \tag{27}$$

and

$$\langle S_0^- S_g^+ \rangle = -m + 2JzC_{11}(1/N) \sum_k \frac{d-\gamma_k}{E_k(2)} \coth\left[\tfrac{1}{2}\beta E_k(2)\right] \exp(-i\vec{k}\cdot\vec{g}) . \tag{28}$$

One finds that in the paramagnetic phase, Eqs. (27) and (28) reduce to the usual isotropic condition $\langle S_0^- S_g^+ \rangle = 2\langle S_0^z S_g^z \rangle$ for $D/J = 0$.

Expanding the coth $(\tfrac{1}{2}\beta E_k)$ terms according to the relation

$$\tfrac{1}{2}\beta E_k \coth(\tfrac{1}{2}\beta E_k) = 1 + \sum_{n=1}^{\infty} (-1)^{n-1} \frac{B_n(\beta E_k)^{2n}}{(2n)!} , \tag{29}$$

where B_n's are Bernoulli numbers we finally get

$$\langle S_0^z S_g^z \rangle = \frac{\tau}{2} \frac{1}{N} \sum_k \left(\frac{1}{\eta} - \gamma_k\right)^{-1} \exp(-i\vec{k}\cdot\vec{g}) + \frac{1}{6} \frac{S_1}{\tau} \frac{1}{N} \sum_k (1-\gamma_k) \exp(-i\vec{k}\cdot\vec{g}) - O(\tau^{-3}) \tag{30}$$

and

$$\langle S_0^- S_g^+ \rangle = -m + \tau \frac{1}{N} \sum_k \left[\frac{d-\gamma_k}{\alpha - \beta'\gamma_k + \gamma_k^2}\right] \exp(-i\vec{k}\cdot\vec{g}) + \frac{1}{3} \frac{C_{11}}{\tau} \frac{1}{N} \sum_k (d-\gamma_k) \exp(-i\vec{k}\cdot\vec{g}) - O(\tau^{-3}) , \tag{31}$$

where $\tau = K_B T/zJ$.

Taking the limit $m \to 0$, utilizing the approximation (25), and considering the vector $\vec{g}$ equal to [000], [100], [110], and [200] for a simple-cubic lattice, we get the following set of coupled equations from Eqs. (30) and (31):

$$C_0 = \tfrac{1}{2}\tau_C I_0(\eta) + S_1/(6\tau_C) , \tag{32}$$

$$C_1 = \tfrac{1}{2}\tau_C I_8(\eta) - S_1/(6z\tau_C) , \tag{33}$$

$$C_2 = \tfrac{1}{2}\tau_C I_{110}(\eta) , \tag{34}$$

$$C_4 = \tfrac{1}{2}\tau_C I_{200}(\eta) , \tag{35}$$

$$S_0 = [AI_0(1) + BI_0(p)]\tau_C + C_{11}/(3\tau_C) , \tag{36}$$

$$S_1 = [AI_8(1) + BI_8(p)]\tau_C - C_{11}/(3z\tau_C) , \tag{37}$$

$$S_2 = [AI_{110}(1) + BI_{110}(p)]\tau_C , \tag{38}$$

$$S_4 = [AI_{200}(1) + BI_{200}(p)]\tau_C , \tag{39}$$

where

$$A = (d-1)/(\alpha-1) , \tag{40}$$

$$B = 1 - A , \tag{41}$$

$$p = 1/\alpha , \tag{42}$$

and $I_g(p)$'s are the extended Watson sums[16,17]

$$I_g(p) = \frac{1}{N} \sum_k \left[\frac{1}{p} - \gamma_k\right]^{-1} \exp(-i\vec{k}\cdot\vec{g}) . \tag{43}$$

Above equations along with the relations

$$C_0 + S_0 = S(S+1) , \tag{44}$$

$$1/\mu^1 = (S_0 + S_4 + 4S_2)/(zS_1) , \tag{45}$$

$$1/\mu = [C_0 + C_4 + 4C_2 + \tfrac{1}{2}(S_0 + S_4 + 4S_2)]/(zC_{11}) , \tag{46}$$

are solved self-consistently for various values of spin and D/J values and the results for a spin-1 simple-cubic lattice is shown in Fig. 1.
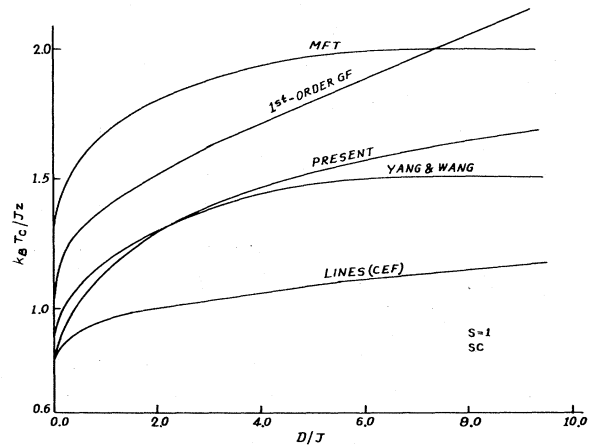

FIG. 1. Present result for T_C is plotted against D/J for a spin-1, simple-cubic lattice, along with the results of some earlier authors.

IV. CONCLUDING REMARKS

In Fig. 1 we have shown, along with the present results, the results obtained from the molecular-field theory, first-order RPA Green's-function theory, correlated-effective-field theory,[18] and from the Green's-function diagram method of Yang and Wang.[8] The figure shows that present results are impressively close to those of Yang and Wang. The agreement for small values of D/J are relatively better than those for large D/J, and it is fully consistent with the approximations $A = 0$ and $B = 1$ (valid for small D/J) adopted in carrying out the computations.

The sensitivity of the Curie temperature to J/D is also examined and it is found that the limiting value of $k_B T_C/zJ$ for $J/D \rightarrow 0$ is very large compared to that obtained in the diagram method. This discrepancy is also due to our approximation mentioned above. In spite of this limitation it may be admitted that the present theory has got the spirit of reproducing better results and one can expect that some better approximations will endorse more accuracy.

It will be relevant here to mention some features of a second-order Green's-function theory in comparison to that of a first-order one. Calculations in the earlier sections show that the energy spectra $E_k(1)$ and $E_k(2)$ are independent of the spontaneous magnetization m, and hence the theory predicts the existence of spin-wave-like excitations well into the paramagnetic phase. On the other hand, first-order Green's-function theories with similar decoupling approximations give energy spectra containing m, and hence according to these theories all sorts of long- and short-range orderings should vanish at the transition temperature, which is contrary to the experimental observations.[19,20] Moreover, due to the presence of m in the energy spectra, the higher-order terms of coth $(\frac{1}{2}\beta E_k)$ expansion of these theories vanish at T_C, and hence one has to include all the aspects of spin correlation in the first term and as a consequence the decoupling approximations assume complicated forms. Whereas, in a second-order theory the spin correlations appear as algebraic equations and hence a simple decoupling like RPA is expected to give good results.

We are, however, unable to present a complete discussion of the low-temperature phase at this stage due to severe complicacies of the involved quantities. We shall treat them in our future discussions of the problem.

ACKNOWLEDGMENTS

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