Thermodynamic properties of Heisenberg magnetic systems*

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In this paper, we present a comprehensive investigation of the effects of the transverse correlation function (TCF) on the thermodynamic properties of Heisenberg antiferromagnetic (AFM) and ferromagnetic (FM) systems with cubic lattices. The TCF of an FM system is positive and increases with temperature, while that of an AFM system is negative and decreases with temperature. The TCF lowers internal energy, entropy and specific heat. It always raises the free energy of an FM system but raises that of an AFM system only above a specific temperature when the spin quantum number is $S \geq 1$. Comparisons between the effects of the TCFs on the FM and AFM systems are made where possible.

Keywords: Heisenberg magnetic system, thermodynamic property, transverse correlation function, Green's function method

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1. Introduction

Magnetic systems have been successfully described by the well-known Heisenberg model. This model could be treated by a number of approximation methods such as molecular-field theory,[1,2] the Opechowski high-temperature expansion method,[3,4] the cluster method by Oguchi[5] and the many-body Green's function method (MBGFM) of quantum statistical theory.[6–22] The MBGFM is valid in the whole temperature range, a merit superior to other methods. The treatment of Heisenberg ferromagnetic (FM)[6–9] and antiferromagnetic (AFM) models[10,11] by means of the MBGFM has been proved successful.

Although magnetization, parallel susceptibility, and Néel temperature of Heisenberg AFM systems have been investigated by researchers,[23–37] a comprehensive study of the thermodynamic properties of FM and AFM systems is desired. In this paper, we present such a comprehensive study, focusing on the effects of the transverse correlation function (TCF) which measures the correlation between two spins in directions perpendicular to the magnetization. We first derive the internal energies of AFM and FM systems by means of the MBGFM including the contribution from the TCF, and then the thermodynamics quantities of FM and AFM systems, such as magnetization, parallel susceptibility, internal energy, entropy, and specific heat, are numerically calculated in detail.

The TCF of an FM system is positive and increases with temperature, while that of an AFM system is negative and decreases with temperature. Specially, the TCF of an FM system vanishes at zero temperature but that of an AFM does not. The TCF has a considerable contribution to the thermodynamics properties. It lowers internal energy, entropy, and specific heat. It always raises the free energy of an FM system but only above a specific temperature for an AFM system when the spin quantum number is $S \geq 1$.

The thermodynamics properties of an AFM system in the presence of an external magnetic field are also investigated. Under the small fields we considered, they have an unremarkable contribution to the thermodynamics properties below the Néel point. Above this point, the increase in the internal energy, entropy and decrease in the specific heat with temperature are observable compared to those without the TCF, while the free energy remains decreasing with temperature in succession.

This paper is organized as follows. Section 2 derives the formulas of internal energies of AFM and FM systems. Section 3 investigates numerically the magnetization, parallel magnetic susceptibility, TCF, and its effects on thermodynamic properties. Section 4 gives our conclusions.

2. Formulas of internal energy

We first derive the formulas of the internal energy of an AFM system, and then present that of an FM system consequently. An AFM lattice is considered to be divided into two sublattices denoted by subscripts 1 and 2. When an external magnetic field is present, the spins in sublattices 1 and 2 are parallel and antiparallel to the field, respectively, the former referred to as the spin-up sublattice. After the formula of internal energy of an AFM system is achieved, we can easily obtain that of an FM system by simply letting the two sublattices be identical.

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The Heisenberg Hamiltonian of an AFM lattice is

$$\begin{aligned} H &= \frac{J}{2}\sum_{(i,j)} \boldsymbol{S}_{i1}\cdot\boldsymbol{S}_{j2} - B_z\sum_i (S^z_{i1}+S^z_{i2}) \\ &= \frac{J}{2}\sum_{(i,j)}\left[\frac{1}{2}(S^+_{i1}S^-_{j2}+S^-_{i1}S^+_{j2})+S^z_{i1}S^z_{j2}\right] \\ &\quad - B_z\sum_{\mu=1}^{2}\sum_i S^z_{i\mu}, \end{aligned} \tag{1}$$

where $J>0$ is the exchange parameter and (i,j) means that only the nearest-neighboring (NN) exchange is considered. $\boldsymbol{S}_{i\mu}=(S^x_{i\mu},S^y_{i\mu},S^z_{i\mu})$ represents the spin operator at site i in the sublattice μ and $S^{\pm}_{i\mu}=S^x_{i\mu}\pm \mathrm{i}S^y_{i\mu}$, B_z is an external magnetic field along the z direction. The Heisenberg exchange in the first term includes two parts: the transverse and longitudinal correlations between spins $\boldsymbol{S}_{i1}$ and $\boldsymbol{S}_{i2}$. The second term is the Zeeman energy under the external field. Since the AFM lattice consists of two sublattices, the Hamiltonian is applicable to both simple cubic (sc) and body-center cubic (bcc) lattices. This is an isotropic exchange model and it has the spontaneous magnetization in the three-dimensional case.[38] In this paper, we set $\mu_{\mathrm{B}}=1$ and $k_{\mathrm{B}}=1$. The internal energy is the statistical average of the Hamiltonian. The internal energy per lattice site is $E_{\mathrm{AFM}}=E_{\mathrm{C,AFM}}+E_{\mathrm{T,AFM}}$, where

$$E_{\mathrm{C,AFM}} = \frac{J}{2N}\sum_{(i,j)}\langle S^z_{i1}S^z_{j2}\rangle - \frac{B_z}{2N}\sum_{\mu=1}^{2}\sum_i\langle S^z_{i\mu}\rangle, \tag{2}$$

$$E_{\mathrm{T,AFM}} = \frac{J}{2N}\sum_{(i,j)}\frac{1}{2}\langle S^+_{i1}S^-_{j2}+S^-_{i1}S^+_{j2}\rangle. \tag{3}$$

Here we use $\langle\ \rangle$ to denote the statistical average, and z_0 is the number of nearest neighbors of one lattice site. Under the mean field approximation, equation (2) can be simplified into $E_{\mathrm{C,AFM}}=J_0\langle S^z_1\rangle\langle S^z_2\rangle/2-B_z\sum_{\mu=1}^{2}\langle S^z_\mu\rangle/2$. The other part concerns the TCF and should be treated carefully. We use the Green's function method to calculate the TCF.

The retarded Green's function is defined as[39]

$$\begin{aligned} G(t-t') &= \langle\langle A(t);B(t')\rangle\rangle \\ &= -\mathrm{i}\theta(t-t')\langle A(t)B(t')-B(t')A(t)\rangle. \end{aligned} \tag{4}$$

In order to calculate the TCF between the two spins located in different sublattices, $\langle S^+_{i1}S^-_{j2}\rangle$, the operator A is taken as $A=S^+_{m1},S^+_{j2}$ and the operator $B=\mathrm{e}^{uS^z_{n1}}S^-_{n1}$. The method of the equation of motion of the Green's functions is used.[40] In decoupling the higher order Green's functions into the lower order ones, the random phase approximation is made. Then the Green's functions are Fourier-transformed and we obtain

$$\begin{aligned} \omega g_{11}(\boldsymbol{k},\omega) &= 2\langle S^z_1\rangle + J_{\boldsymbol{k}}\langle S^z_1\rangle g_{21}(\boldsymbol{k},\omega) \\ &\quad + (-J_0\langle S^z_2\rangle + B_z)g_{11}(\boldsymbol{k},\omega), \end{aligned} \tag{5}$$

$$\begin{aligned} \omega g_{21}(\boldsymbol{k},\omega) &= J_{\boldsymbol{k}}\langle S^z_2\rangle g_{11}(\boldsymbol{k},\omega) \\ &\quad + (-J_0\langle S^z_1\rangle + B_z)g_{21}(\boldsymbol{k},\omega), \end{aligned} \tag{6}$$

where functions g are the Fourier transformation of G, $J_{\boldsymbol{k}}=J\sum_j \mathrm{e}^{\mathrm{i}\boldsymbol{k}\cdot(j-m)}$, $J_0=z_0J$ and the summation over j covers the nearest neighbors of site m. The function $g_{11}(\boldsymbol{k},\omega)$ is a propagator within sublattice 1 and $g_{21}(\boldsymbol{k},\omega)$ is another one between the two sublattices. By exchanging the subscripts 1 and 2, we obtain the expressions of $g_{22}(\boldsymbol{k},\omega)$ and $g_{12}(\boldsymbol{k},\omega)$. The four equations constitute a set of linear equations $[\omega\boldsymbol{I}-\boldsymbol{P}]\boldsymbol{g}=\boldsymbol{F}$, where $\boldsymbol{I}$ is the identity matrix and $\boldsymbol{g}$ is the Green's function matrix.[14,41] $\boldsymbol{F}$ is a commutator matrix of operators A and B

$$\begin{aligned} \boldsymbol{F} &= \begin{pmatrix} \langle[S^+_1,\mathrm{e}^{uS^z_1}S^-_1]\rangle & 0 \\ 0 & \langle[S^+_2,\mathrm{e}^{uS^z_2}S^-_2]\rangle \end{pmatrix} \\ &= \{\{(\mathrm{e}^{-u}-1)[S_\mu(S_\mu+1)\psi_\mu(u)+\psi'_\mu(u)-\psi''_\mu(u)] \\ &\quad + 2\psi'_\mu(u)\}\delta_{\mu\nu}\}, \end{aligned} \tag{7}$$

where $\psi_\mu(u)=\langle\mathrm{e}^{uS^z_\mu}\rangle$, $\boldsymbol{P}$ is termed as the Hamiltonian matrix, the elements of which are determined by Eqs. (5) and (6), $\boldsymbol{U}$ is the eigenvector matrix of $\boldsymbol{P}$ given by

$$\boldsymbol{U} = \begin{pmatrix} \dfrac{J_{\boldsymbol{k}}\langle S^z_1\rangle}{\sqrt{J^2_{\boldsymbol{k}}\langle S^z_1\rangle^2+(J_0\langle S^z_2\rangle-B_z+\omega_1)^2}} & \dfrac{J_{\boldsymbol{k}}\langle S^z_1\rangle}{\sqrt{J^2_{\boldsymbol{k}}\langle S^z_1\rangle^2+(J_0\langle S^z_2\rangle-B_z+\omega_2)^2}} \\ \dfrac{J_0\langle S^z_2\rangle-B_z+\omega_1}{\sqrt{J^2_{\boldsymbol{k}}\langle S^z_1\rangle^2+(J_0\langle S^z_2\rangle-B_z+\omega_1)^2}} & \dfrac{J_0\langle S^z_2\rangle-B_z+\omega_1}{\sqrt{J^2_{\boldsymbol{k}}\langle S^z_1\rangle^2+(J_0\langle S^z_2\rangle-B_z+\omega_2)^2}} \end{pmatrix} \tag{8}$$

and the eigenvalues of $\boldsymbol{P}$ are just the spin-wave elementary excitation spectra (EES) of the system

$$\begin{aligned} \omega_{1,2}(\boldsymbol{k}) &= \frac{1}{2}J_0\{-\langle S^z_1\rangle-\langle S^z_2\rangle\pm[(\langle S^z_1\rangle-\langle S^z_2\rangle)^2 \\ &\quad + 4\gamma^2_{\boldsymbol{k}}\langle S^z_1\rangle\langle S^z_2\rangle]^{1/2}\}+B_z, \end{aligned} \tag{9}$$

where $\gamma_{\boldsymbol{k}}=J_{\boldsymbol{k}}/J_0$.

The well-known spectral theorem allows one to calculate the statistical average of the product of the operators $\langle BA\rangle$[40,42]

$$\begin{aligned} \langle B_{n\nu}A_{m\mu}\rangle &= \frac{2}{N}\sum_{\boldsymbol{k}}\sum_{\tau}\frac{U_{\mu\tau}U^{-1}_{\tau\nu}}{\mathrm{e}^{\beta\omega_\tau}-1}F_\nu\,\mathrm{e}^{-\mathrm{i}\boldsymbol{k}\cdot(m-n)} \\ &= \Phi_{m\mu,n\nu}F_\nu, \end{aligned} \tag{10}$$

where the quantities $\Phi_{m\mu,n\nu}$ have been defined.

We take $n = m$ and $\mu = \nu$ in Eq. (10) to obtain the diagonal elements of the correlation functions $\langle B_\mu A_\mu \rangle = \Phi_\mu F_\mu$, where

$$\Phi_\mu = \frac{2}{N}\sum_{k}\sum_{\tau}\frac{U_{\mu\tau}U^{-1}_{\tau\mu}}{e^{\beta\omega_\tau}-1}.$$

The function $\psi_\mu(u) = \langle e^{uS^z_\mu}\rangle$ observes an ordinary differential equation of the second order, from which the expression of the sublattice magnetization can be solved[9,43]

$$\langle S^z_\mu\rangle = \frac{(\Phi_\mu+S_\mu+1)\Phi_\mu^{2S_\mu+1}-(\Phi_\mu-S_\mu)(\Phi_\mu+1)^{2S_\mu+1}}{(\Phi_\mu+1)^{2S_\mu+1}-\Phi_\mu^{2S_\mu+1}}. \tag{11}$$

When taking $u = 0$ in Eq. (7), we obtain from Eq. (10) the TCF of the AFM system

$$K_{\rm AFM} \equiv \langle S^+_{m\mu}S^-_{n\nu}\rangle = 2\Phi_{m\mu,n\nu}\langle S^z_\nu\rangle. \tag{12}$$

By multiplying exchange J and making summation over the nearest neighbors of site m, we further obtain the corresponding transverse correlation energy (TCE) $\Theta_{\rm AFM} \equiv J\sum_n\langle S^+_{m\mu}S^-_{n\nu}\rangle = 2\Xi_{\mu\nu}\langle S^z_\nu\rangle$, where

$$\Xi_{\mu\nu} = \frac{2}{N}\sum_{k}\sum_{\tau}\frac{U_{\mu\tau}U^{-1}_{\tau\nu}}{e^{\beta\omega_\tau}-1}J_k.$$

Thus it follows from Eq. (3) that the TCE per site is $E_{\rm T,AFM} = (\Xi_{12}\langle S^z_2\rangle + \Xi_{21}\langle S^z_1\rangle)/2$. In the absence of the external field, the two sublattices are of space-reversal symmetry, so that it must be $\Xi_{12}\langle S^z_2\rangle = \Xi_{21}\langle S^z_1\rangle$. This is easily confirmed. When $B_z = 0$, it is derived that

$$\Phi_1(T) = \frac{2}{N}\sum_{k}\frac{\coth[\beta J_0(1-\gamma_k^2)^{1/2}/2]}{2(1-\gamma_k^2)^{1/2}} - \frac{1}{2} \tag{13}$$

and $\Phi_2 = -\Phi_1 - 1$. Consequently, one achieves $\langle S^z_1\rangle = -\langle S^z_2\rangle = \langle S^z\rangle_{\rm AFM}$ from Eq. (11). On the other hand, it is derived that

$$\Xi_{21}(T) = -\frac{2}{N}\sum_{k}\frac{J_0\gamma_k^2\coth[\beta\langle S^z\rangle_{\rm AFM}J_0(1-\gamma_k^2)^{1/2}/2]}{2(1-\gamma_k^2)^{1/2}}, \tag{14}$$

and $\Xi_{12} = -\Xi_{21}$. Therefore, we have $\Xi_{12}\langle S^z_2\rangle = \Xi_{21}\langle S^z_1\rangle$.

The values of the TCF at zero temperature and the Néel point $T_{\rm N}$ are of particular interest. As $T = 0$,

$$\Phi_1(0) = \frac{2}{NJ_0}\sum_{k}\frac{1}{2(1-\gamma_k^2)} - \frac{1}{2},$$
$$\Xi_{21}(0) = -\frac{2}{N}\sum_{k}\frac{J_k\gamma_k}{2(1-\gamma_k^2)^{1/2}}.$$

As $T = T_{\rm N}$, it can be derived from Eq. (10) that the TCF and TCE of the AFM system are

$$K_{\rm AFM}(T_{\rm N}) = -\frac{4T_{\rm N}}{NJ_0}\sum_{k}\frac{\gamma_k}{1-\gamma_k^2}e^{-i\boldsymbol{k}\cdot(\boldsymbol{m}-\boldsymbol{n})}, \tag{15}$$

$$\Theta_{\rm AFM}(T_{\rm N}) = -\frac{4T_{\rm N}}{N}\sum_{k}\frac{\gamma_k^2}{1-\gamma_k^2}. \tag{16}$$

If J is replaced by $-J$, the AFM system will become an FM system and $\langle S^z_1\rangle = \langle S^z_2\rangle = \langle S^z\rangle_{\rm FM}$. The total internal energy per spin of the FM system is written as $E_{\rm FM} = E_{\rm C,FM} + E_{\rm T,FM}$, where $E_{\rm C,FM} = -zJ_0(\langle S^z\rangle_{\rm FM})^2/2 - B_z\langle S^z\rangle_{\rm FM}$ is the internal energy of the FM system without the TCF. The TCE is

$$E_{\rm T,FM} = -\frac{\langle S^z\rangle_{\rm FM}}{N}\sum_{k}\frac{J_k}{e^{\beta\omega_2}-1}.$$

The magnetization is

$$\langle S^z\rangle_{\rm FM} = \frac{(\Psi_2+S+1)\Psi_2^{2S+1}-(\Psi_2-S)(\Psi_2+1)^{2S+1}}{(\Psi_2+1)^{2S+1}-\Psi_2^{2S+1}}, \tag{17}$$

where

$$\Psi_2 = \frac{1}{N}\sum_{k}\frac{1}{e^{\beta\omega_2}-1}.$$

The spin-wave EES of the FM lattice becomes $\omega_{1,2} = (J_0 \pm J_k)\langle S^z\rangle_{\rm FM} + B_z$. Because the lattice is now divided into two sublattices, the first Brillouin zone of FM crystal is collapsed to a half to induce an optical branch, which is the reason that a sign "+" appears in the EES. In the absence of the external field, one easily obtains from Eq. (15) that $\langle S^z\rangle_{\rm FM}(0) = S$, and as a result, the TCF of FM system $K_{\rm FM} = 0$. This means that at 0 K, there is no quantum fluctuation in the FM system. At the Curie temperature $T_{\rm C}$, the TCF of the FM system is

$$K_{\rm FM}(T_{\rm C}) = \langle S^+_m S^-_n\rangle = \frac{4T_{\rm C}}{NJ_0}\sum_{k}\frac{\gamma_k}{1-\gamma_k^2}e^{-i\boldsymbol{k}\cdot(\boldsymbol{m}-\boldsymbol{n})} \tag{18}$$

and the corresponding TCE is

$$\Theta_{\rm FM}(T_{\rm C}) = -J\sum_{n}\langle S^-_m S^+_n\rangle = -\frac{4T_{\rm C}}{N}\sum_{k}\frac{\gamma_k^2}{1-\gamma_k^2}. \tag{19}$$

Under the random phase approximation

$$T_{\rm N} = T_{\rm C} = \frac{J_0S(S+1)}{3R}, \tag{20}$$

where

$$R = \frac{2}{N}\sum_{k}\frac{1}{1-\gamma_k}.$$

Comparisons of Eqs. (15) and (16) with Eqs. (18) and (19) lead to

$$K_{\rm FM}(T_{\rm C}) = -K_{\rm AFM}(T_{\rm N}), \tag{21}$$

$$\Theta_{\rm FM}(T_{\rm C}) = \Theta_{\rm AFM}(T_{\rm N}). \tag{22}$$

That is to say, at the order–disorder phase transition temperature, the TCFs of the AFM and FM systems with the same lattice structure are contrary numbers of each other, and their TCEs are the same. When equation (20) is applied into Eqs. (15), (16), (18), and (19), it is seen that the functions $K_{\rm AFM}(T_{\rm N})/S(S+1)$, $\Theta_{\rm AFM}(T_{\rm N})/S(S+1)$, $K_{\rm FM}(T_{\rm C})/S(S+1)$, and $\Theta_{\rm FM}(T_{\rm C})/S(S+1)$ are independent of the spin quantum number.

3. Results and discussion

We investigate the thermodynamic properties of both AFM and FM systems in detail. The magnetization, parallel susceptibility, internal energy, free energy, entropy, and specific heat of both systems with sc and bcc lattices are calculated. In numerical calculation, J is set to be 100. The sublattice magnetizations of AFM systems versus temperature are plotted in Fig. 1 for several parameters. Figures 1(a) and 1(b) plot the sublattice magnetization versus temperature curves at several spin quantum numbers of AFM systems with sc and bcc lattices. At 0 K, the two sublattice magnetizations are not rigorously antiparallel to each other. As a result, the z-direction magnetizations are less than the spin quantum numbers. It was the well-known zero-point spin deviation and a quantum effect, which could not be eliminated by applying an external magnetic field or other means.[42,44,45]

Figures 1(c) and 1(d) depict the sublattice magnetizations $\langle S_1^z\rangle$ and total magnetizations $\langle S^z\rangle_{\rm T} = \langle S_1^z\rangle + \langle S_2^z\rangle$ of sc lattice at $S = 1/2$ under several external fields. The total magnetizations are always zero at 0 K, and at finite temperature they increase with either the field or temperature. The reason is that the exchange interaction enforces the spins of the two sublattices to align in an antiparallel way but thermal motion counteracts partly the exchange interaction to make the spins more easily align in the field direction.

The susceptibility is the response of a material to an external magnetic field. In this paper, we merely concentrate on the zero-field parallel susceptibility. It follows from Eq. (11) that

$$\chi_\mu = \frac{{\rm d}\langle S_\mu^z\rangle}{{\rm d}B_z} = \frac{{\rm d}\Phi_\mu}{{\rm d}B_z}\left\{\frac{(2S_\mu+1)^2\Phi_\mu^{2S_\mu}(\Phi_\mu+1)^{2S_\mu}}{[(\Phi_\mu+1)^{2S_\mu+1}-\Phi_\mu^{2S_\mu+1}]^2}-1\right\}. \quad (23)$$

The response to a small magnetic field is linear and the linear response is the same for both sublattices,[42] such that the AFM magnetization can be expressed as $\langle S_\mu^z\rangle = \pm\langle S^z\rangle_0 + \chi_0 B_z$, where $\langle S^z\rangle_0$ is the spontaneous magnetization and χ_0 is the parallel susceptibility. Above the Néel point, the spontaneous magnetization vanishes. As $B_z \to 0$ and $\Phi_\mu \to \infty$, $\langle S_\mu^z\rangle \to S(S+1)/3\Phi_\mu$ and ${\rm e}^{\beta\omega_\mu} - 1 \to \beta\omega_\mu$. We obtain

$$\chi_{0,\rm AFM}^{-1} = \frac{3T}{S(S+1)}\frac{2}{N}\sum_{\boldsymbol{k}}\frac{1-J_0\chi_{0,\rm AFM}}{(1-J_0\chi_{0,\rm AFM})^2 - J_{\boldsymbol{k}}^2\chi_{0,\rm AFM}^2}. \quad (24)$$

At the Néel point, because of $(6T_{\rm N}/NJ_0)\sum_{\boldsymbol{k}}(1/1-\gamma_{\boldsymbol{k}}^2) = S(S+1)$, the susceptibility reaches its maximum $\chi_{0,\rm AFM}(T_{\rm N}) = 1/J_0$. Similarly, starting from Eq. (17), we obtain the formula evaluating the susceptibility of an FM system when the temperature is above the Curie point

$$\chi_{0,\rm FM}^{-1} = \frac{3T}{NS(S+1)}\sum_{\boldsymbol{k}}\frac{1}{\chi_{0,\rm FM}J_0(1-\gamma_{\boldsymbol{k}})+1}. \quad (25)$$

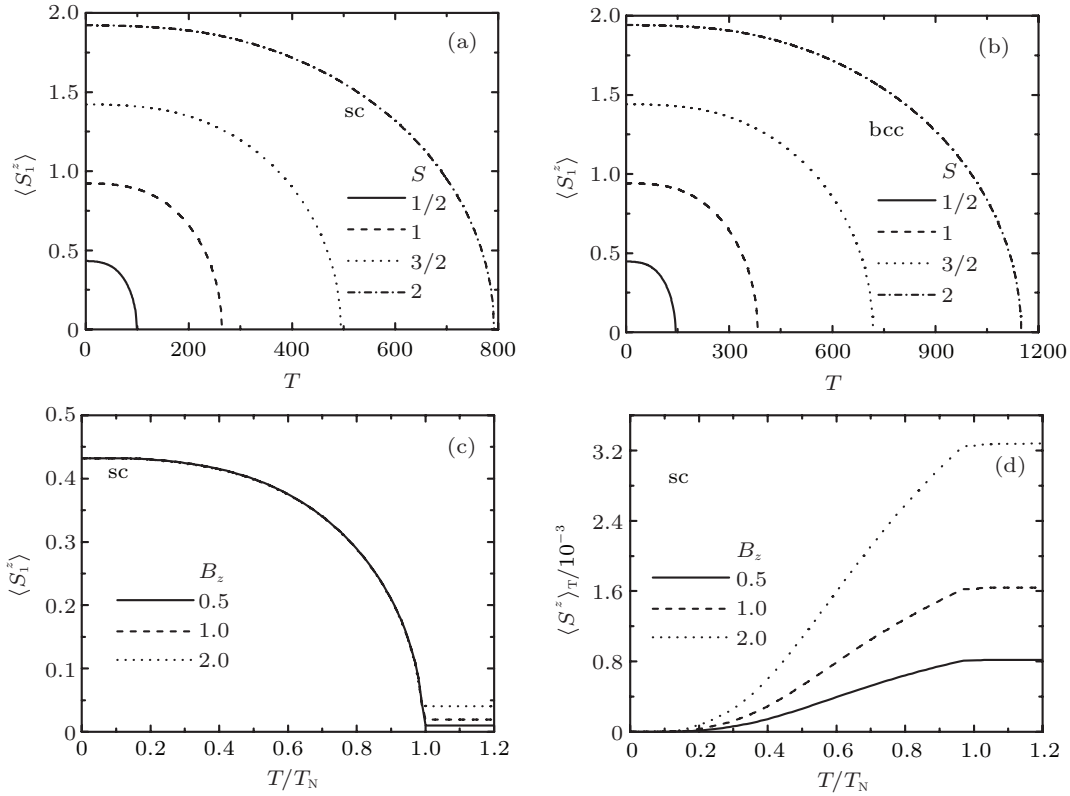


Fig. 1. Sublattice magnetizations of AFM systems vs. temperature curves at several parameters. Panels (a) and (b) are sublattice magnetizations when $B_z = 0$. (c) Sublattice magnetizations and (d) the total magnetizations $\langle S^z\rangle_{\rm T} = \langle S_1^z\rangle + \langle S_2^z\rangle$ of sc lattice at $S = 1/2$ under several magnetic fields.

Figure 2(a) plots the susceptibilities of FM systems. They are infinity at the Curie point, and decrease with temperature above the Curie point for the systems becoming the paramagnetic state. The susceptibilities of AFM systems are plotted in Figs. 2(b) and 2(c). They are zero at 0 K and increase with temperature until the Néel point. Above this point, they decrease with temperature, a behavior similar to the FM systems. The remarkable difference between the FM and AFM systems appears at their order–disorder transition points. The total spontaneous magnetizations of the AFM systems are always zero near the Néel point, so that the susceptibilities at the Néel point are finite. While the FM spontaneous magnetizations are nonzero near the Curie point, thus the susceptibilities are infinite.

The TCF between spins in the two sublattices describes the correlation between spins in directions perpendicular to the magnetization and plays an important role on the total internal energy. Figure 3(a) plots the TCFs of FM and AFM systems. The TCFs of FM systems are positive and increase with temperature, while those of the AFM systems are negative and decrease with temperature. At zero temperature, $K_{\rm FM}(0)=0$, while $K_{\rm AFM}(0)<0$. At the order–disorder transition point, both the terms $K_{\rm FM}(T_{\rm C})/S(S+1)$ and $K_{\rm AFM}(T_{\rm N})/S(S+1)$ are indeed independent of spin quantum number S, as mentioned at the end of the last section. The numerical results in Fig. 3 verify Eq. (21). Figure 3(b) plots the TCEs of FM and AFM systems. They are negative and decrease with temperature increasing. At zero point, $E_{\rm T,FM}(0)=0$ while $E_{\rm T,AFM}(0)<0$. At the order–disorder transition point, both the terms $E_{\rm T,FM}(T_{\rm C})/S(S+1)$ and $E_{\rm T,AFM}(T_{\rm N})/S(S+1)$ are independent of spin quantum number S, also mentioned at the end of the last section. At the order–disorder transition point, the magnetizations of the AFM and FM systems are zero, so that $E_{\rm C,AFM}(T_{\rm N})=0$ and $E_{\rm C,FM}(T_{\rm C})=0$. Then, it follows from Eq. (22) that $E_{\rm T,FM}(T_{\rm C})=E_{\rm T,AFM}(T_{\rm N})$, as reflected in Fig. 3(b). Figure 3(c) plots the TCEs at zero temperature vs. spin quantum number S. The FM TCE is always zero while the AFM TCE is negative and increases with spin quantum number S.

The free energy is expressed by $F=E-TS_{\rm E}$,[46] where we have used $S_{\rm E}$ to denote the entropy. Both the free energy and entropy can be calculated in terms of the internal energy

$$F(T)=E(0)-T\int_0^T\frac{E(T')-E(0)}{T'^2}\,{\rm d}T', \tag{26}$$

$$S_{\rm E}(T)=\frac{E(T)-E(0)}{T}+\int_0^T\frac{E(T')-E(0)}{T'^2}\,{\rm d}T'. \tag{27}$$

The specific heat is $C_V(T)=(\partial E(T)/\partial T)_V$. In Fig. 4 the internal energies, free energies, entropies, and specific heats as functions of temperature for FM systems with sc lattices are plotted by solid lines. The quantities with a subscript c and plotted by dashed lines mean the results where the TCFs are omitted.

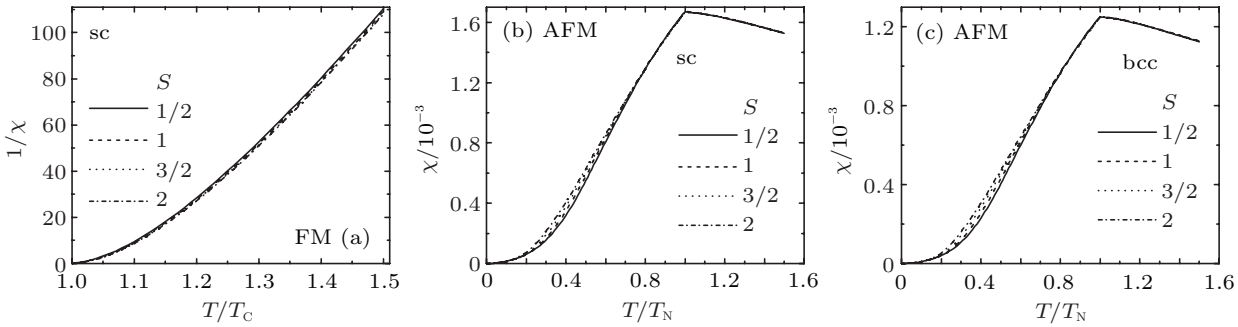


Fig. 2. Parallel susceptibilities of FM and AFM systems. (a) The FM susceptibilities of sc lattices above the Curie point. The AFM susceptibilities of (b) sc and (c) bcc lattices.

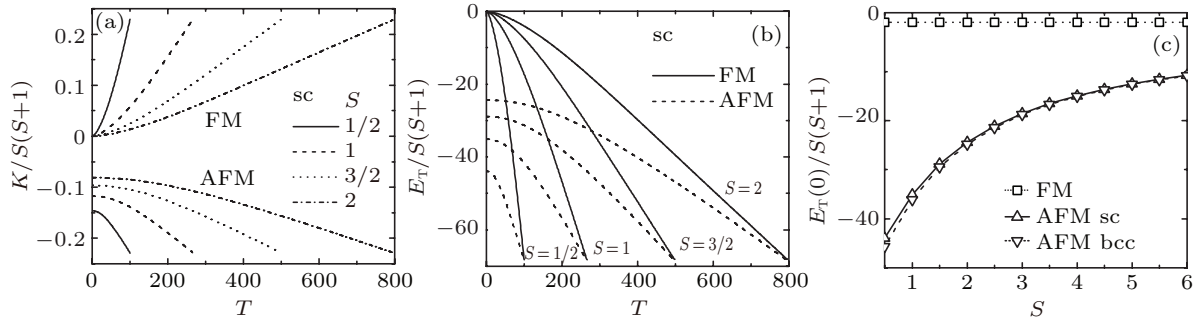


Fig. 3. The TCFs and TCEs of FM and AFM systems when $B_z=0$. (a) $K/S(S+1)$ vs. T for sc lattices, (b) $E_{\rm T}/S(S+1)$ vs. T for sc lattices and (c) $E_{\rm T}(0)/S(S+1)$ vs. S.

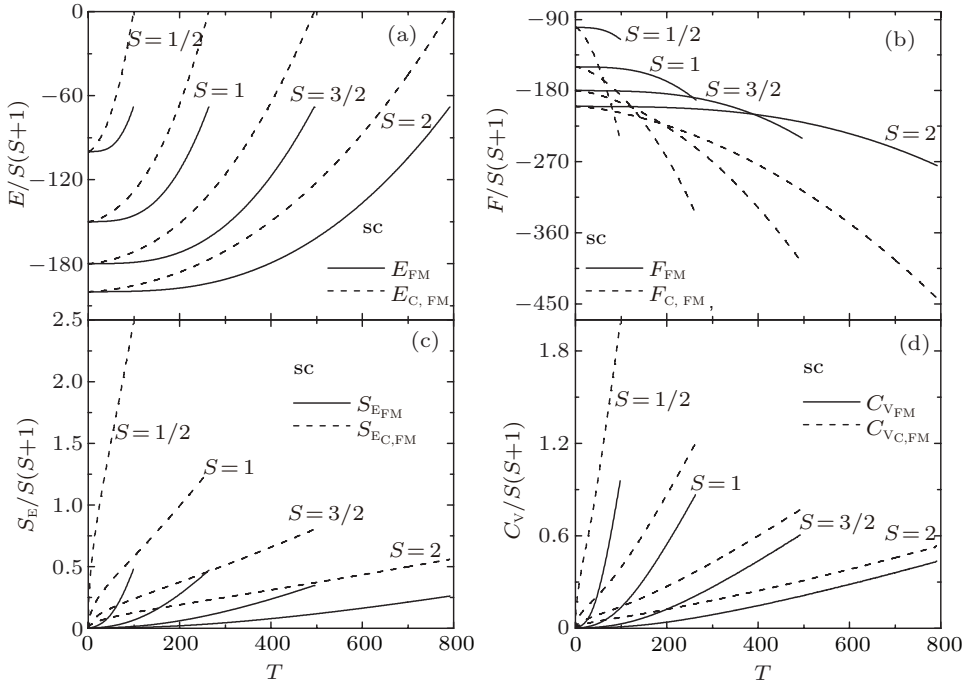


Fig. 4. The thermodynamic quantities of FM sc lattices vs. temperature. (a) Internal energies, (b) free energies, (c) entropies, and (d) specific heats.

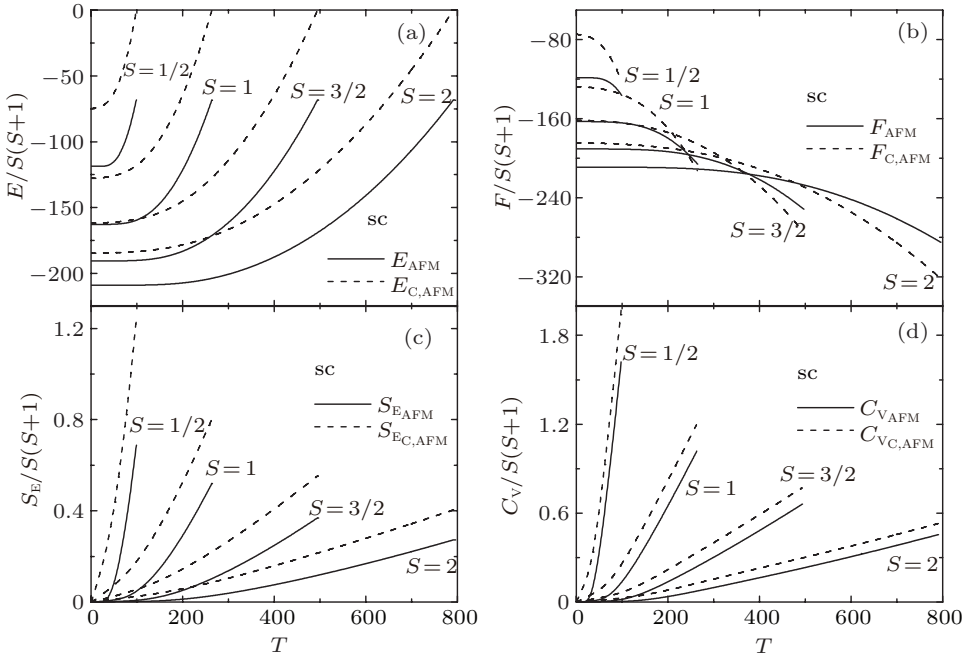


Fig. 5. The thermodynamic quantities of AFM sc lattices vs. temperature. (a) Internal energies, (b) free energies, (c) entropies, and (d) specific heats.

In this way, we are able to clearly see the effect of the transverse correlation. Figure 4(a) plots the internal energies E_{FM} and $E_{C,FM}$. Since the TCFs depress the thermal motion, they always lower the internal energies at finite temperature. At 0 K, the TCFs are zero, so that $E_{FM}(0) = E_{C,FM}(0)$. At the Curie point, $E_{C,FM}(T_C) = 0$, which means that there is no interaction between spins when the transverse effect is neglected. The TCFs raise the free energies and slow down their decrease with temperature, as shown in Fig. 4(b). In Figs. 4(c) and 4(d), it is seen that the increase of the entropy and specific heat is depressed observably by the TCFs. A uniform feature of Fig. 4 is that all the solid lines vary not so steeply as the corresponding dashed lines. This means that the TCFs more or less counteract the variation of thermodynamic quantities.

In Fig. 5, the four thermodynamic quantities of the AFM systems with sc lattices are plotted. Again, the solid and dashed lines present the results with and without transverse correlation effect, respectively. A first impression of Fig. 5 is the same as that of Fig. 4: all the solid lines vary but not so steeply as the corresponding dashed lines. Figure 5(a) shows

that $E_{\rm AFM}(0) < E_{\rm C,AFM}(0)$, demonstrating that the transverse correlation effect lowers the internal energy even at zero temperature. This is a feature that is different from the FM systems. Correspondingly, the free energy is lowered starting from zero temperature when the TCFs are considered, as seen in Fig. 5(b). However, when $S \geq 1$, because the free energy $F_{\rm AFM}$ varies too slowly compared to $F_{\rm C,AFM}$, it becomes larger than the latter above specific temperature values, which is another remarkable feature. The entropies and specific heats are always lowered, which is similar to the cases of the FM systems.

Under the random phase approximation, we are unable to evaluate the thermodynamic quantities for zero sublattice magnetizations when the temperature is above the $T_{\rm N}$. When an external field is applied, the sublattice magnetizations become nonzero as $T > T_{\rm N}$, and we are able to reckon the physical quantities above $T_{\rm N}$. The effects of the external field on the thermodynamic quantities of AFM systems are shown in Fig. 6. Under small fields, the change in these thermodynamics quantities below the Néel point is unremarkable. Above this point, $E_{\rm C,AFM}$ remains almost unchanged while $E_{\rm AFM}$ observably increase with temperature, as shown in Fig. 6(a). In Fig. 5(b), the free energy curves $F_{\rm C,AFM}$ and $F_{\rm AFM}$ of $S = 1/2$ do not meet under the Néel point, while they meet above the Néel point when the field is applied, as seen in Fig. 6(b). Figure 6(c) shows that the behaviors of entropies $S_{\rm EAFM}$ and $S_{\rm EC,AFM}$ are analogous to the energies: $S_{\rm EC,AFM}$ remains almost unvaried above the $T_{\rm N}$ in the presence of the field, while $S_{\rm EAFM}$ still increases above the $T_{\rm N}$ under the field. Figure 6(d) shows that above the Néel point, $C_{V\rm C,AFM}$ is zero because $E_{\rm C,AFM}$ is. While under the field, $C_{V\rm AFM}$ is nozero and decreases with temperature above the $T_{\rm N}$.

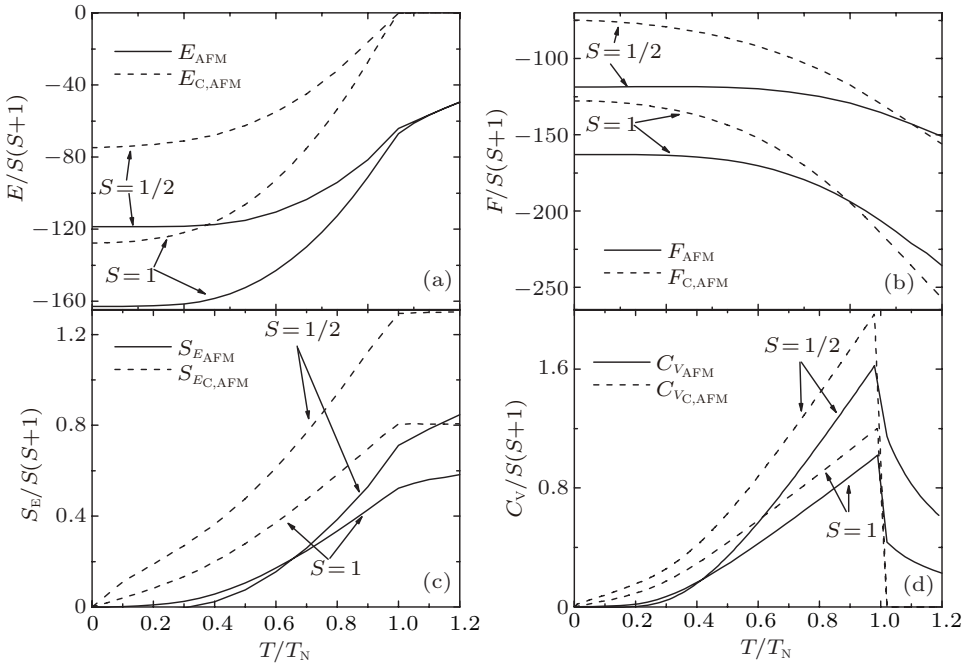


Fig. 6. The thermodynamic quantities of AFM sc lattices for $B_z = 2$ at $S = 1/2$ and 1. (a) Internal energies, (b) free energies, (c) entropies, and (d) specific heats.

4. Concluding remarks

In this paper, we present a comprehensive investigation of the thermodynamic properties of FM and AFM Heisenberg systems, when the transverse correlation effect is included. The expression of the internal energy, including the TCEs, of three-dimensional AFM systems consisting of two sublattices is derived by means of MBGFM. That of an FM system is achieved as a special case where the two sublattices become identical. The thermodynamic quantities, including magnetization, parallel susceptibility, internal energy, free energy, entropy, and specific heat of both FM and AFM systems are investigated numerically in detail. Particular attention is paid to the effect of the TCF. The TCF of an AFM system is nonzero at zero point and decreases with temperature, while that of an FM system is zero and increases with temperature. The TCEs arising from the transverse correlation effect are negative and decrease with temperature. On the whole, the TCF has a considerable effect on the thermodynamics quantities. It lowers the internal energy, entropy, and specific heat for both FM and AFM systems. For FM systems, the TCF always raises the free energy, while for AFM systems, it is not until a specific temperature. That is to say, the curves $F_{\rm C,AFM}$ and $F_{\rm AFM}$ of an AFM system have a cross. For $S \geq 1$, the cross is at a temperature below $T_{\rm N}$, and for $S = 1/2$ it is above $T_{\rm N}$. A small external magnetic field does not influence the thermodynamic quantities much, while it enables one to calculate the quantities above the Néel point so as to give the correct temperature dependence of them: the internal energy and entropy increase

with and specific heat decreases with temperature. Finally, the TCFs more or less counteract the variation of thermodynamic quantities with temperature, so that the curves vary not so steeply as those without including the TCFs.

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