The sublattice magnetizations of the spin-$s$ quantum Heisenberg antiferromagnets

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We study the spin-$s$ quantum Heisenberg antiferromagnetic models in a magnon theory free of any unphysical magnon state. Because the unphysical magnon states are completely removed in the magnon Hamiltonians and during the approximation process, we derive spin-$s$ ($s > \frac{1}{2}$) reliable Néel temperature $T_N$ and reasonable sublattice magnetization unified for $T \leq T_N$.

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Quantum Heisenberg antiferromagnetic (QHAFM) models are well-accepted models for insulating antiferromagnets. The insulating cuprates $La_2CuO_4$ and $RBa_2Cu_3O_6$ ($R$ is an rare earth element) and their light-doped samples are typical antiferromagnets of spin $1/2$.\[3\] Because of their proximity to the high temperature superconductivity in the doped cuprates, they have absorbed very much attention. On the other hand, insulating magnetic materials of high spin, such as the well-known Manganese materials, recently become important.\[2\] They can be described by the QHAFM models. But, for the QHAFM models, there has not yet existed any reasonable spin-$s$ ($s > \frac{1}{2}$) Néel temperature $T_N$ and sublattice magnetization unified for all temperature $T \leq T_N$.

On the other hand, the defining spin operators in the QHAFM models are neither Bose nor Fermi operators. Many authors tried to transform the spin operators into the standard Bose (magnon) operators since Bloch invented the concept of spin wave.\[1\] On the experimental side authors tend to make use of spin wave (magnon) theory to explain their experimental results.\[3\] They can be described by the QHAFM models. But, for the QHAFM models, there has not yet existed any reasonable spin-$s$ ($s > \frac{1}{2}$) Néel temperature $T_N$ and sublattice magnetization unified for all temperature $T \leq T_N$.

Therefore, magnon (or spin wave) theory is a very powerful approach to the QHAFM models. In this approach one makes use of Holstein-Primakoff\[5\] or Dyson\[6\] transformation to transform the original spin Hamiltonians into magnon Hamiltonians. On a single site, the magnon state space is infinitely dimensional, but there are only $2s+1$ physical spin states. There exist infinite extra unphysical magnon states in any magnon Hamiltonian. In removing effect of the unphysical states, many methods were proposed,\[1\][2][3] but the problem of the unphysical magnon states remained unsolved. In a magnon theory free of the unphysical states,\[3\] all of the unphysical states were put infinitely high in energy and their effect was removed in the magnon Hamiltonians and during the approximation process. In the case of spin $1/2$, reliable ferromagnetic magnetization and antiferromagnetic sublattice magnetization were obtained under a simple but sensible approximation.

In this Letter we study the ($s \geq 1$) quantum Heisenberg antiferromagnetic models in the magnon theory free of the unphysical states. In the way similar to those in the previous works,\[3\] we remove effect of the unphysical magnon states in the magnon Hamiltonians and during the approximation process, and thereby derive for the first time reliable analytical Néel temperature $T_N$ and sublattice magnetization, being reasonable from zero to $T_N$, of the quantum Heisenberg antiferromagnetic model of any spin ($s \geq 1$) in three dimensions. The zero-temperature normalized sublattice magnetization $M_0/s$ increases monotonously with spin $s$. The reduced sublattice magnetization $M/M_0$ decreases monotonously with spin $s$ increasing at any given reduced temperature $T/T_N$ between 0 and 1. Our sublattice magnetizations are substantial improvements on those of the existing nonlinear spin wave (magnon) theories, which became unreasonable at high temperature. Our Néel temperature in the spin-$s$ case is only 4% larger than the series expansion result.\[4\]

The standard spin-$s$ QHAFM Hamiltonian is defined by

$$H = \sum_{\langle ij \rangle} J_{ij} \left[ \frac{1}{2} (S_i^+ S_j^- + S_i^- S_j^+) + S_i^z S_j^z \right]. \quad (1)$$

The summation is over the nearest neighbor sites of the lattice. The exchange constant $J_{ij}$ is positive and, in general, is direction dependent to describe real space anisotropy. We suppose our lattice can be divided into two sublattices so that the nearest neighbors of any site of one sublattice belong to the other sublattice. Making a $\pi$ rotation in the spin space of one of the two sublattices, the Hamiltonian (1) becomes

$$H = \sum_{\langle ij \rangle} J_{ij} \left[ \frac{1}{2} (S_i^+ S_j^+ + S_i^- S_j^-) - S_i^z S_j^z \right]. \quad (2)$$

To transform the spin operators into the magnon operators, We choose Holstein-Primakoff (HP) transformation:

$$S_i^- = a_i^\dagger \sqrt{2s - n_i}, \quad S_i^+ = \sqrt{2s - n_i} a_i, \quad S_i^z = s - n_i, \quad (3)$$

where the magnon number operator $n_i$ is defined by $n_i = a_i^\dagger a_i$. The magnon operators $a$ are standard Bose operators. After substituting the magnon transformation (3) into the Hamiltonian (2), we obtain the HP magnon Hamiltonian:
\[
H_m = \sum_{\langle ij \rangle} J_{ij} \left[ \frac{1}{2} (a^\dagger_i a^\dagger_j A_i A_j + \text{h.c.}) - n_in_j \right] + \sum_i \epsilon a^\dagger_i a_i + H_0
\]

where \( H_0 = -\frac{1}{2} JZs^2N, \epsilon = JZs, \) and \( A_i = \sqrt{2s-n_i} \).

The magnon Hamiltonian is different from the original spin Hamiltonian because the magnon operator introduces an infinite unphysical magnon states, so we only \( \sum \) the terms of at most 1 magnon Green functions to construct some composite functions. The equation of motion of operator \( X \) is defined by

\[
\frac{d}{dt} \langle \langle X \rangle \rangle = \sum_i J_i \langle \langle T_i \rangle \rangle
\]

and

\[
\frac{d}{dt} \langle \langle X \rangle \rangle = \sum_i (z - \gamma) \langle \langle B_i \rangle \rangle + \sum_i J_i \langle \langle T_i \rangle \rangle
\]

The magnon square root \( A_i = \sqrt{2s-n_i} \) in the Hamiltonian can be expanded in terms of \( a_i^m a_i^\dagger_m \). The \( U \) term makes the unphysical site correlation \( \langle a_i^m a_i^\dagger_m \rangle \) (\( m \geq 2s+1 \)) exactly equivalent to zero. So that the expansion of \( \sqrt{2s-n_i} \) is accurately truncated into a sum of \( 2s+1 \) magnon operator product terms:

\[
A_i = \sum_{m=0}^{2s} (-1)^m C_m \frac{1}{m!} a_i^m a_i^\dagger_m
\]

\[
C_m = \sum_i (-1)^i \frac{m!}{i!(m-i)!} \sqrt{2s-i}.
\]

Substituting the \( A_i \) expression into the magnon Hamiltonian, \( H_U \), our total Hamiltonian becomes a finite sum of rational magnon operator products. Therefore, the infinite-\( U \) term \( H_U \) removes effect of the unphysical magnon states and at the same time simplify the magnon Hamiltonian.\[13\]

Being similar to the previous work,\[13\] we work in the framework of Green function method and the approach of equation of motion. We define \( \langle \langle X \rangle \rangle \) to be the Green function of the operators \( X \) and \( Y \) in Zubarev notation. The equation of motion of operator \( X \) is defined to be \( \frac{d}{dt} \langle \langle X \rangle \rangle = \hat{X} \) or \( z \langle \langle X \rangle \rangle = \hat{X} \langle \langle X \rangle \rangle \) in the frequency (\( z \)) space. Since the magnon number is not conserved for the AFM models, we need two kinds of Green functions. Intuitively, we have \( \langle \langle a_i a_j^\dagger \rangle \rangle \) and \( \langle \langle a_i^\dagger a_j \rangle \rangle \). But they cannot compose a closed set of equations of motion because of the infinite \( U \) term in the magnon Hamiltonian. On-site multimagnon Green functions inevitably appear due to the \( U \) term. We need a series of the on-site multimagnon Green functions to construct some composite Green functions so that their equations of motion are closed and contain terms of at most \( 1/U \) order.\[13\] For this reason, we construct the two series of Green functions: \( \langle \langle B_i | D^m_k \rangle \rangle \) and \( \langle \langle B^\dagger_i | D^m_k \rangle \rangle \) (\( m = 1, 2, 3, \ldots, 2s \)), where \( D^m_j = a_j^m a_j^{m-1} \) and \( B_i = A_i a_i \). Here the operator \( A_i \) is equivalent to \( A_i \) with the \( a_i^\dagger a_i^\dagger \) term removed. The Green functions satisfy the equations of motion

\[
(z - \epsilon) \langle \langle B_i | D^m_j \rangle \rangle = W^m_i \delta_{ij} + \sum_l J_{il} \langle \langle T_{il} | D^m_j \rangle \rangle
\]

and

\[
(z + \epsilon) \langle \langle B^\dagger_i | D^m_j \rangle \rangle = W^m_i \delta_{ij} - \sum_l J_{il} \langle \langle T^\dagger_{il} | D^m_j \rangle \rangle,
\]

where \( W^m_i = \langle \langle A_i a_i^\dagger A_i^\dagger | D^m_i \rangle \rangle \), \( W^m_i = \langle \langle a_i^\dagger A_i^\dagger | D^m_i \rangle \rangle \), and \( T_{il} = (s-n) a_i^\dagger A_i - n A_i a_l \). In Eqs. \[8\] and \[9\], other terms of \( 1/U \) order or smaller terms are neglected since they are zero when \( U \) tends to infinity. We make the on-site approximation \( n_i X_i = \langle \langle n_i | X_i = \langle \langle n_i | X \rangle \rangle \) for the operator \( n_i X_i \) only if \( i \neq l \). Under this approximation, the equations of motion reduce to

\[
(z - \gamma) \langle \langle B_i | D^m_j \rangle \rangle = W^m_i \delta_{ij} + \sum_l J_{il} (s-n) \langle \langle B_i | D^m_j \rangle \rangle
\]

and

\[
(z + \gamma) \langle \langle B^\dagger_i | D^m_j \rangle \rangle = -\sum_l J_{il} (s-n) \langle \langle B_i | D^m_j \rangle \rangle,
\]

where \( \gamma = JZ(s-n) \). The parameter \( J \) is the largest one of the exchange \( J_{ij} \) and the effective coordination number \( \sum \) is defined by \( Z = \sum_{j(i)} J_{ij}/J \), where the \( j \) summation is made over all nearest neighbor sites of site \( i \). It is not difficult to solve the above equations by transforming the Green functions into the \( k \)-space. The resultant Green functions in the \( k \)-space read:

\[
\langle \langle B_k | D^m_k \rangle \rangle = \frac{W^m [z + JZ(s-n)]}{z^2 - J^2 Z^2(s-n)^2(1 - r_k^2)}
\]

and

\[
\langle \langle B^\dagger_k | D^m_k \rangle \rangle = \frac{-W^m JZ(s-n)r_k}{z^2 - J^2 Z^2(s-n)^2(1 - r_k^2)}
\]

The spectrum function \( r_k \) is defined by \( r_k = (2JZ) \sum_{j(i)} J_{il} \exp(iR_l - iR_j) \). It is obvious that the magnon correlation functions \( \langle \langle D^m_k B_k \rangle \rangle = V^m_i \) and \( W^m_i \) both are site independent. Therefore we obtain the self-consistent equations

\[
V^m = W^m \psi
\]

and

\[
\psi = \frac{1}{N} \sum_k \left[ \frac{1}{1 - r_k} \coth \left( \frac{\beta \gamma}{2} \sqrt{1 - r_k^2} \right) - 1 \right].
\]
For spin \( s \), we have only \( 2s \) non-zero on-site correlation functions: \( p_n = \langle P_n \rangle (1 \leq n \leq 2s) \), where the on-site multimagnon operator \( P_n \) is defined by \( P_n = \sum a_n^\dagger a_\zeta^\dagger a_\eta a_n \). The unphysical correlation functions \( P_n (n \geq 2s + 1) \) are exactly equivalent to zero. The self-consistent equation (13) can be solved by

\[
p_n = \frac{\psi^n}{(1 + \psi)^{2s+1} - \psi^{2s+1}} \sum_{k=0}^{2s-n} C_k^{2s+1} \psi^k,
\]

where \( n \leq 2s \) and the coefficient \( C_m^n = n!/[m!(n-m)!] \) is the binomial coefficient. Since \( S_\zeta^2 = s - n \) and \( n = p_1 \), we derive the following spin magnetization:

\[
\langle S^2 \rangle = s - \psi + \frac{(2s+1)\psi^{2s+1}}{(1 + \psi)^{2s+1} - \psi^{2s+1}}.
\]

At very low temperature, \( \psi \) can be expanded as \( \psi = \psi_0 + cT^2 \) (\( c \) is a positive constant.), where \( \psi_0 = (\xi - 1)/2 \) and \( \xi = \frac{1}{N} \sum k 1/\sqrt{1 - r_k^2} \). As a result, we derive \( \langle S^2 \rangle = S_0^2 - c_1T^2 \) (\( c_1 \) is also a positive constant.). \( S_0^2 \) is given by the right-hand side of formula (17) with \( \psi \) replaced by \( \psi_0 \). The Néel temperature \( T_N \) is given by

\[
T_N = \frac{JZ}{3s\eta}(s + 1),
\]

where the constant \( \eta \) is given by \( \eta = \frac{1}{N} \sum k 1/(1 - r_k^2) \).

When temperature approaches to the Néel temperature \( T_N \), the sublattice magnetization \( \langle S^2 \rangle \) is given by the asymptotic expression:

\[
\langle S^2 \rangle = \alpha \sqrt{1 - \frac{T}{T_N}}.
\]

The positive constant \( \alpha \) is defined by \( \alpha = \frac{2}{s(s + 1)[1 + \frac{2}{s} + \frac{2}{s}(2s^2 + 2s - 9)]^{1/2}} \). In one dimension, the integrations \( \eta \) and \( \xi \) both diverge. There is no antiferromagnetic order at any temperature. In two dimensions the integration \( \xi \) converges, but the integration \( \eta \) diverges. There is antiferromagnetic order only at zero temperature. These are consistent with the Mermin-Wagner theorem. [5]

At general temperature, one cannot derive any analytical expression. We present some digital results in three-dimensional (3D) simple-cubic lattice. In Fig. 1 we show the sublattice magnetizations of spins 1/2, 1, 3/2, and 2 as functions of temperature. For convenience, we normalize the saturation magnetizations into one, that is, we divide the magnetizations by the corresponding spin \( s \). The normalized sublattice magnetizations increase with spin \( s \) increasing at zero temperature. For comparison with the results of the existing spin wave (magnon) theories, we also present the sublattice magnetizations of a typical version of the existing nonlinear spin wave (magnon) theories (NLSW). It is obvious that the NLSW magnetizations are unreasonable at high temperature, especially near the Néel temperatures. In fact, all of the existing NLSW theories yielded double-valued magnetizations at high temperature. [16][17] Therefore the existing spin wave (magnon) theories work at most at low temperature, and our magnon theory free of unphysical states works well in whole temperature region.

![FIG. 1. Sublattice magnetizations (divided by \( s \)) as functions of temperature (in unit of \( J \)) for spin 1/2 (see Ref 13), 1, 3/2, and 2. The solid lines are the results of the magnon theory free of any unphysical states. The dashed lines are results from other magnon (spin wave) theories (Ref 16). The latter are not reasonable at high temperature (Ref 16, 17).](image)

![FIG. 2. The reduced sublattice magnetizations (\( M/M_0 \)) as functions of reduced temperature (\( T/T_N \)) for spin 1/2 (see Ref 13), 1, 2, 4, 9, and \( \infty \). The reduced sublattice magnetization decreases with reduced temperature increasing for a given spin \( s \). It also decreases with spin \( s \) increasing at a given reduced temperature.](image)
magnetization is linear in the reduced temperature at $T/T_N = t \sim 0$. It is very interesting that the reduced magnetization monotonously decreases with spin $s$ at any given reduced temperature $t$ between 0 and 1.

The above results are obtained under the postulation of isotropy ($J_{ij} = J$) in the 3D simple cubic lattice. In fact, the approach and the on-site approximation can be successfully applied to the cases of anisotropic exchange constants where $J_{ij}$ is direction dependent. In the cases of the cuprates, one has an in-plane exchange constant $J$ and an interplane exchange constant $\delta J$ ($\delta$ is small).

For line-like materials, one has an on-site exchange constant $J$ and an interline constant $\delta J$ with small $\delta$.

The method and approximation process we make use of in this paper are similar to the previous work on the quantum Heisenberg spin-$\frac{1}{2}$ ferromagnetic models and spin-$\frac{1}{2}$ antiferromagnetic model. In those cases, our magnetizations and sublattice magnetization were equivalent to the reasonable results which had been obtained by means of the direct spin-operator methods. In the case of spin $1/2$, we obtain $T_N = 0.988J$ in the simple cubic lattice, being only 4% larger than the series expansion result. Therefore, the reliability of our spin-$s$ sublattice magnetizations and Néel temperatures in this paper can be justified by the equivalence in the ferromagnetic case and the comparison with the accurate series expansion result.

In summary, we have studied the quantum Heisenberg antiferromagnetic models of spin $s$ in a magnon theory free of the unphysical states. Effect of the unphysical states is completely removed in the magnon Hamiltonians and during the approximation process. We thereby derive reliable analytical the Néel temperatures and sublattice magnetizations unified for all temperatures $T \leq T_N$ and obtain the multimagnon correlation functions for any spin. At any given reduced temperature, the reduced sublattice magnetization monotonously decreases with spin $s$ increasing. Our sublattice magnetizations are substantial improvements on those of the existing spin wave (magnon) theories. Our Néel temperature in the case of spin $1/2$ is only 4% larger than the series expansion result.

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