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Modified spin-wave theory of a square-lattice antiferromagnet

Minoru Takahashi
Institute for Solid State Physics, University of Tokyo, 7-22-1 Roppongi, Minato-ku, Tokyo 106, Japan
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A modified spin-wave theory for the Heisenberg antiferromagnet ($\mathcal{H}=J\sum_{\langle ij\rangle}\mathbf{S}_i\cdot\mathbf{S}_j$) is formulated under the assumption of zero sublattice magnetization in the same way with the Heisenberg ferromagnet. This theory gives self-consistent equations which are equivalent to those of Auerbach and Arovas, but in our theory the factor of $\frac{3}{2}$ in the correlation function does not appear. This theory reproduces the main results of traditional spin-wave theory, as well as those of renormalization-group theory, in a unified picture. For the square lattice at low temperature the susceptibility behaves as $a+bT$ and the correlation length as $(c/T)\exp(d/T)$. This correlation length coincides very well with experimental results of La_2CuO_4 if we choose $J=900$ K. Calculation of self-consistent equations is done for the $S=\frac{1}{2}$ system and compared with the result of exact diagonalization of a 4×4 system and high-temperature expansion. The quantitative agreement is surprisingly good, especially at $T\lesssim0.6J$.

I. INTRODUCTION

Recently properties of the Heisenberg antiferromagnet on a square lattice have been investigated by many theorists[1] in connection with the mechanisms of high-T_c superconductivity.[2] In many models, high-T_c superconductors may be regarded as a Heisenberg antiferromagnet with dopant holes. The undoped parent compound La_2CuO_4 (Ref. 3) is known to be an antiferromagnetic insulator. Its behavior may then be described by a Heisenberg Hamiltonian with $S=\frac{1}{2}$ on the square lattice.

In previous papers[4] we proposed a modified spin-wave theory for a Heisenberg ferromagnet in zero magnetic field. We introduced a chemical potential which is determined by the condition of zero magnetization. In one dimension, the results agree very well with those of the thermodynamic Bethe-*Ansatz* integral equation for a 1D ferromagnet. The results for the 2D classical Heisenberg ferromagnet agree very well with Monte Carlo results for this system at low temperature. For the analysis of low-temperature properties, the spin-wave theory is very successful even in the case of no long-range order. So in this paper we try to apply the same method to the antiferromagnetic Heisenberg model, on the square lattice in particular.

In Sec. II Anderson and Kubo's spin-wave theory[5] is reformulated with the condition of zero sublattice magnetization and free energy minimum. Surprisingly we get the Auerbach and Arovas (AA) equations[6] which were obtained by Schwinger boson formulation. In Sec. III the square-lattice Heisenberg antiferromagnet is analyzed at low temperature. For the ground-state energy, specific heat, and elementary excitation, our theory reproduces the results of conventional spin-wave theory.[5] Moreover, our theory determines correlation functions, susceptibility, and correlation length. The results are consistent with one-loop renormalization-group theory. In Sec. IV spin-wave results for energy and susceptibility are compared with high-temperature expansion and exact diagonalization of the $S=\frac{1}{2}$ 4×4 system. Quantitative agreement is good. The correlation length of spin-wave theory is compared with experimental results of La_2CuO_4 and we get excellent agreement. In the Appendix we analyze the classical antiferromagnet and get almost the same equation with classical ferromagnets.

II. FORMULATION OF ANTIFERROMAGNETIC SPIN-WAVE THEORY AT $H=0$

We consider the following Hamiltonian:

$$\mathcal{H}=J\sum_{\langle ij\rangle}(S_i^xS_j^x+S_i^yS_j^y+S_i^zS_j^z)-H\sum_i S_i^z\ , \tag{1a}$$

$$[S_l^\alpha,S_j^\beta]=i\varepsilon_{\alpha\beta\gamma}S_l^\gamma\delta_{lj},\quad S_l^2=S(S+1)\ . \tag{1b}$$

Here $\langle ij\rangle$ means that i and j sites are nearest neighbors. We assume that the lattice is bipartite and divided into A and B sublattices. On the A (B) sublattice, the vacuum state is the $S^z=S(-S)$ state. We introduce the antiferromagnetic Dyson-Maleev (ADM) transformation[7] for $S_l^+\equiv S_l^x+iS_l^y$, $S_l^-\equiv S_l^x-iS_l^y$ and S_l^z instead of Holstein-Primakoff (HP) transformation,[8]

$$S_l^-=a_l^\dagger,\quad S_l^+=(2S-a_l^\dagger a_l)a_l,\quad S_l^z=S-a_l^\dagger a_l\quad \text{for } l\in A\ , \tag{2a}$$

$$S_m^-=-b_m,\quad S_m^+=-b_m^\dagger(2S-b_m^\dagger b_m)\ ,$$
$$S_m^z=-S+b_m^\dagger b_m\quad \text{for } m\in B\ . \tag{2b}$$

Commutation relations (1b) are satisfied by the following relations:

$$[a_l,a_{l'}^\dagger]=\delta_{ll'},\quad [b_m,b_{m'}^\dagger]=\delta_{mm'}$$

and

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$$0=[a_l,a_{l'}]=[b_m,b_{m'}]=[a_l^\dagger,a_{l'}^\dagger]=[b_m^\dagger,b_{m'}^\dagger]=[a_l^\dagger,b_m]=[a_l,b_m^\dagger]=[a_l^\dagger,b_m^\dagger]=[a_l,b_m]\ . \tag{2c}$$

All spin operators are written in four kinds of Bose operators: a, $a^\dagger$, b, and $b^\dagger$. Spin-pair operator $\mathbf{S}_i\cdot\mathbf{S}_j$ are written as follows:

$$\begin{aligned}\mathbf{S}_l\cdot\mathbf{S}_m&=-S^2+S(a_l^\dagger a_l+b_m^\dagger b_m-a_l^\dagger b_m^\dagger-a_lb_m)\\&\quad+\tfrac{1}{2}a_l^\dagger(b_m^\dagger-a_l)^2b_m\ ,\end{aligned}\tag{3a}$$

$l\in A,\ m\in B$

$$\begin{aligned}\mathbf{S}_l\cdot\mathbf{S}_{l'}&=S^2-S(a_l^\dagger-a_{l'}^\dagger)(a_l-a_{l'})\\&\quad-\tfrac{1}{2}a_l^\dagger a_{l'}^\dagger(a_l-a_{l'})^2\ ,\end{aligned}\tag{3b}$$

l and $l'\in A,\ l\neq l'$,

$$\begin{aligned}\mathbf{S}_m\cdot\mathbf{S}_{m'}&=S^2-S(b_m^\dagger-b_{m'}^\dagger)(b_m-b_{m'})\\&\quad-\tfrac{1}{2}(b_m^\dagger-b_{m'}^\dagger)^2b_mb_{m'}\ ,\end{aligned}\tag{3c}$$

m and $m'\in B,\quad m\neq m'$.

Then the Hamiltonian (1a) at $H=0$ is written as follows:

$$\begin{aligned}\mathcal{H}=&-\tfrac{1}{2}JzS^2N+J\sum_{\langle lm\rangle}S(a_l^\dagger a_l+b_m^\dagger b_m-a_l^\dagger b_m^\dagger-a_lb_m)\\&+\tfrac{1}{2}a_l^\dagger(b_m^\dagger-a_l)^2b_m\ ,\end{aligned}\tag{3d}$$

where z is the number of nearest neighbors and N is the total number of sites. If we regard a,b and $a^\dagger,b^\dagger$ as Hermitian conjugate operators, the Hamiltonian (3d) has an infinite number of eigenstates. All eigenstates of (1a) are contained in (3d). It is expected that the ground state is common for both Hamiltonians. We believe that the gap between the lowest unphysical state and the physical ground state is about JS^2. In ADM transformation there is no term higher than fourth order and the Hamiltonian is non-Hermitian. This is different from HP transformation. Next we introduce the ideal spin-wave density matrix with Bogoliubov transformation,

$$\rho=\exp\left[\frac{1}{T}{\sum_{\mathbf{k}}}'\varepsilon_{\mathbf{k}}(\alpha_{\mathbf{k}}^\dagger\alpha_{\mathbf{k}}+\beta_{-\mathbf{k}}^\dagger\beta_{-\mathbf{k}})\right]\ ,\tag{4a}$$

$$\begin{aligned}\alpha_{\mathbf{k}}&=\cosh\theta_{\mathbf{k}}\,a_{\mathbf{k}}-\sinh\theta_{\mathbf{k}}\,b_{-\mathbf{k}}^\dagger\ ,\\\beta_{-\mathbf{k}}^\dagger&=-\sinh\theta_{\mathbf{k}}\,a_{\mathbf{k}}+\cosh\theta_{\mathbf{k}}\,b_{-\mathbf{k}}^\dagger\ .\end{aligned}\tag{4b}$$

Here ${\sum}'_{\mathbf{k}}$ means the sum of $\mathbf{k}$ over half of the first Brillouin zone, $a_{\mathbf{k}}$ and $b_{-\mathbf{k}}^\dagger$ are the Fourier transforms of a_l and $b_m^\dagger$,

$$\begin{aligned}a_{\mathbf{k}}&=\sqrt{2/N}\sum_{l\in A}a_l\exp(-i\mathbf{k}\cdot\mathbf{r}_l)\ ,\\b_{-\mathbf{k}}^\dagger&=\sqrt{2/N}\sum_{m\in B}b_m^\dagger\exp(-i\mathbf{k}\cdot\mathbf{r}_m)\ .\end{aligned}\tag{4c}$$

For the density matrix (4) we have

$$\begin{aligned}\langle a_l^\dagger b_m\rangle=\langle a_lb_m^\dagger\rangle=\langle a_l^\dagger a_{l'}^\dagger\rangle=\langle a_la_{l'}\rangle=\langle b_m^\dagger b_{m'}^\dagger\rangle\\=\langle b_mb_{m'}\rangle=0\ ,\end{aligned}\tag{5a}$$

$$\begin{aligned}\langle a_l^\dagger a_{l'}\rangle&=f(\mathbf{r}_l-\mathbf{r}_{l'})-\tfrac{1}{2}\delta_{ll'}\ ,\\\langle b_m^\dagger b_{m'}\rangle&=f(\mathbf{r}_m-\mathbf{r}_{m'})-\tfrac{1}{2}\delta_{mm'}\ ,\\\langle a_l^\dagger b_m^\dagger\rangle&=\langle a_lb_m\rangle=g(\mathbf{r}_l-\mathbf{r}_m)\ ,\end{aligned}\tag{5b}$$

$$\begin{aligned}f(\mathbf{r})&\equiv\frac{2}{N}{\sum_{\mathbf{k}}}'\cosh(2\theta_{\mathbf{k}})\exp(-i\mathbf{k}\cdot\mathbf{r})(\tilde{n}_{\mathbf{k}}+\tfrac{1}{2})\ ,\\g(\mathbf{r})&\equiv\frac{2}{N}{\sum_{\mathbf{k}}}'\sinh(2\theta_{\mathbf{k}})\exp(-i\mathbf{k}\cdot\mathbf{r})(\tilde{n}_{\mathbf{k}}+\tfrac{1}{2})\ ,\end{aligned}\tag{5c}$$

$$\tilde{n}_{\mathbf{k}}\equiv\langle\alpha_{\mathbf{k}}^\dagger\alpha_{\mathbf{k}}\rangle=\langle\beta_{\mathbf{k}}^\dagger\beta_{\mathbf{k}}\rangle=[\exp(\varepsilon_{\mathbf{k}}/T)-1]^{-1}\ .\tag{5d}$$

Using Wick's theorem[9] we have the expectation value of spin pairs in Eqs. (3a)–(3c),

$$\begin{aligned}&\langle\mathbf{S}_l\cdot\mathbf{S}_m\rangle=-[S+\tfrac{1}{2}-f(\mathbf{0})+g(\mathbf{r}_l-\mathbf{r}_m)]^2\ ,\\&l\in A\ \text{and}\ m\in B\end{aligned}\tag{6a}$$

$$\begin{aligned}&\langle\mathbf{S}_l\cdot\mathbf{S}_{l'}\rangle=[S+\tfrac{1}{2}-f(\mathbf{0})+f(\mathbf{r}_l-\mathbf{r}_{l'})]^2\ ,\\&l,l'\in A\ \text{or}\ l,l'\in B,\ l\neq l'\ .\end{aligned}\tag{6b}$$

At $H=0$ the magnetization of each site should be zero

$$0=\langle S_l^z\rangle=S+\tfrac{1}{2}-f(\mathbf{0})\ .\tag{7}$$

Energy is given by

$$\mathcal{E}=-(JN/2)\sum_{\boldsymbol{\delta}}[S+\tfrac{1}{2}-f(\mathbf{0})+g(\boldsymbol{\delta})]^2\ ,$$

where $\boldsymbol{\delta}$'s are vectors to the nearest neighbors. Entropy is

$$\mathcal{S}=2{\sum_{\mathbf{k}}}'\{(\tilde{n}_{\mathbf{k}}+1)\ln(\tilde{n}_{\mathbf{k}}+1)-\tilde{n}_{\mathbf{k}}\ln\tilde{n}_{\mathbf{k}}\}\ .$$

We should minimize $F=\mathcal{E}-T\mathcal{S}$ under the condition (7). Using

$$\partial[F/N-\mu f(\mathbf{0})]/\partial\theta_{\mathbf{k}}=0$$

and

$$\partial[F/N-\mu f(\mathbf{0})]/\partial\varepsilon_{\mathbf{k}}=0$$

we have

$$\begin{aligned}&\tanh(2\theta_{\mathbf{k}})=\eta\gamma_{\mathbf{k}}\ ,\\&\varepsilon_{\mathbf{k}}=\lambda[\cosh(2\theta_{\mathbf{k}})-\eta\gamma_{\mathbf{k}}\sinh(2\theta_{\mathbf{k}})]\ ,\end{aligned}\tag{8a}$$

$$\begin{aligned}&\lambda\equiv Jzg(\boldsymbol{\delta})-\mu,\quad\eta\equiv Jzg(\boldsymbol{\delta})/\lambda\ ,\\&\gamma_{\mathbf{k}}\equiv z^{-1}\sum_{\boldsymbol{\delta}}\exp(i\mathbf{k}\cdot\boldsymbol{\delta})\ .\end{aligned}\tag{8b}$$

Here μ is a Lagrange multiplier, or the chemical potential. We assumed that all $g(\boldsymbol{\delta})$'s are the same. From Eq. (8a) we have

$$\varepsilon_{\mathbf{k}}=\lambda(1-\eta^2\gamma_{\mathbf{k}}^2)^{1/2},\ \cosh(2\theta_{\mathbf{k}})=1/(1-\eta^2\gamma_{\mathbf{k}}^2)^{1/2}\ ,$$
$$\sinh(2\theta_{\mathbf{k}})=\eta\gamma_{\mathbf{k}}/(1-\eta^2\gamma_{\mathbf{k}}^2)^{1/2}\ . \tag{8c}$$

Thus we have a set of self-consistent equations for η and λ from (7) and (8),

$$S+\frac{1}{2}=\frac{2}{N}\sum_{\mathbf{k}}{}'\frac{1}{(1-\eta^2\gamma_{\mathbf{k}}^2)^{1/2}}\frac{1}{2}\times\coth\left[\frac{\lambda}{2T}(1-\eta^2\gamma_{\mathbf{k}}^2)^{1/2}\right]\ . \tag{9a}$$

$$\frac{\eta\lambda}{Jz}=\frac{2}{N}\sum_{\mathbf{k}}{}'\frac{\eta\gamma_{\mathbf{k}}^2}{(1-\eta^2\gamma_{\mathbf{k}}^2)^{1/2}}\frac{1}{2}\times\coth\left[\frac{\lambda}{2T}(1-\eta^2\gamma_{k}^2)^{1/2}\right]\ . \tag{9b}$$

One trivial solution of these is

$$\eta=g(\delta)=E=0,\quad \lambda=T\ln(1+S^{-1})\ ,$$
$$F=-TN\ln[(1+S)(1+S^{-1})^S]\ . \tag{10}$$

At high temperature this gives minimum free energy. But at low temperature the solution of nonzero η has the minimum free energy. At a certain temperature there should be a jump from the $\eta=0$ solution to the $\eta\neq 0$ solution. Of course this first-order phase transition is an artifact of our approximation. For the $S=\frac{1}{2}$ square lattice, this transition occurs at $T=0.91J$.

From (8c) we find $f(\mathbf{r})=0$ for $(-1)^{\mathbf{r}}=-1$ and $g(\mathbf{r})=0$ for $(-1)^{\mathbf{r}}=1$. Then Eqs. (6a), (6b), and (7) yield

$$\langle \mathbf{S}_i\cdot\mathbf{S}_j\rangle=f^2(\mathbf{r}_i-\mathbf{r}_j)-\tfrac{1}{4}\delta_{i,j}-g^2(\mathbf{r}_i-\mathbf{r}_j)\ . \tag{11}$$

Fourier transform $S(\mathbf{q})$ of the two-point function (11) is as follows:

$$S(\mathbf{q})=\sum_{\mathbf{r}}\exp(i\mathbf{q}\cdot\mathbf{r})\langle \mathbf{S}_0\cdot\mathbf{S}_{\mathbf{r}}\rangle$$
$$=-\frac{1}{4}+\frac{2}{N}\sum_{\mathbf{k}}{}'\cosh(2\theta_{\mathbf{k}+\mathbf{q}}-2\theta_{\mathbf{k}})\times(\tilde{n}_{\mathbf{k}+\mathbf{q}}+\tfrac{1}{2})(\tilde{n}_{\mathbf{k}}+\tfrac{1}{2})\ . \tag{12}$$

Static uniform susceptibility is given by $S(\mathbf{0})$,

$$\chi=\frac{1}{T}\sum_{\mathbf{r}}\langle S_0^zS_{\mathbf{r}}^z\rangle=\frac{1}{3T}S(\mathbf{0})=\frac{1}{3T}\left[\frac{2}{N}\right]\sum_{\mathbf{k}}{}'(\tilde{n}_{\mathbf{k}}^2+\tilde{n}_{\mathbf{k}})\ . \tag{13}$$

$S(\mathbf{K})$ is given by

$$S(\mathbf{K})=\frac{2}{N}\sum_{\mathbf{k}}{}'\frac{1+\eta^2\gamma_{\mathbf{k}}^2}{1-\eta^2\gamma_{\mathbf{k}}^2}(\tilde{n}_{\mathbf{k}}+\tfrac{1}{2})^2-\tfrac{1}{4}\ . \tag{14}$$

Almost equivalent equations were obtained by AA.[6] But in their formulation the right hand side (rhs) of (11) is multiplied by $\frac{3}{2}$. In a recent paper Hirsch and Tang[10] obtained Eqs. (5)–(14) at $T=0$ ($\tilde{n}_{\mathbf{k}}=0$) using HP transformation.

III. INFINITE SQUARE LATTICE

We define state density function $w(x)$

$$w(x)=\frac{2}{N}\sum_{\mathbf{k}}\delta(x-\gamma_{\mathbf{k}}),\quad 0\le x\le 1\ . \tag{15}$$

For a two-dimensional square lattice at $N\to\infty$ we have

$$w(x)=\left[\frac{2}{\pi}\right]^2K((1-x^2)^{1/2})=\frac{2}{\pi}+O(1-x)\ , \tag{16}$$

where $K(k)$ is a complete elliptic integral of the first kind with modulus k. As is well known $w(x)$ has logarithmic singularity at $x=0$.

At $T=0$, η is $1-O(N^{-1})$ and $\tilde{n}_{\mathbf{k}}$ is zero. Then $f(\mathbf{r})$ in (5c) is written as follows:

$$f(\mathbf{r})=\frac{1}{N}(1-\eta^2)^{-1/2}+\tfrac{1}{2}\int_0^{2\pi}\int_0^{2\pi}d\mathbf{k}\frac{1}{(2\pi)^2}(1-\gamma_{\mathbf{k}}^2)^{-1/2}e^{i\mathbf{k}\cdot\mathbf{r}}\ ,$$
$$\text{if } (-1)^{\mathbf{r}}=1\ . \tag{17}$$

Equation (9a) yields

$$\frac{1}{N}(1-\eta^2)^{-1/2}=m_0\equiv S+\tfrac{1}{2}-\tfrac{1}{2}\int_0^1(1-x^2)^{-1/2}w(x)ds$$
$$=S-0.196\,60\ . \tag{18}$$

Asymptotic behavior of (17) for large r at $(-1)^{\mathbf{r}}=1$ is

$$f(\mathbf{r})\simeq m_0+(2\pi)^{-2}\int_0^\infty dk\,k\int_0^{2\pi}d\theta\frac{\sqrt{2}}{k}e^{ikr\cos\theta}$$
$$=m_0+(\sqrt{2}\pi r)^{-1}\ .$$

In the same way we have $g(\mathbf{r})\simeq m_0+(\sqrt{2}\pi r)^{-1}$ at $(-1)^{\mathbf{r}}=-1$. Asymptotic behavior of the two-point function (11) is as follows:

$$\langle \mathbf{S}_0\cdot\mathbf{S}_{\mathbf{r}}\rangle\simeq(-1)^{\mathbf{r}}[m_0+(\sqrt{2}\pi r)^{-1}]^2\ . \tag{19}$$

So m_0 is the spontaneous order of this system. This coincides with Huse's argument[11] which states $1/r$ decay of $(-1)^{\mathbf{r}}\langle \mathbf{S}_0\cdot\mathbf{S}_{\mathbf{r}}\rangle-m_0^2$ at $T=0$. Equation (9b) becomes as follows:

$$\frac{\lambda}{4J}=m_0+\tfrac{1}{2}\int_0^1x^2(1-x^2)^{-1/2}w(x)dx=m_1\ ,$$
$$m_1\equiv S+\tfrac{1}{2}-\tfrac{1}{2}\int_0^1(1-x^2)^{1/2}w(x)dx$$
$$=S+0.078\,974\ . \tag{20}$$

Ground-state energy per site is given by $-2Jm_1^2$. Elementary excitation at $T=0$ given in (8c) is

$$\varepsilon_{\mathbf{k}}=4Jm_1(1-\gamma_{\mathbf{k}}^2)^{1/2}$$

and spin-wave velocity is

$$v=2\sqrt{2}Jm_1\ . \tag{21}$$

These results were already obtained in the usual spin-wave theory.[5]

Next we analyze the low-temperature properties of Eqs. (9). Using state density function (9a) and (9a)$-\eta$(9b) are

$$S+\frac{1}{2}=\frac{1}{2}\int_0^1(1-\eta^2x^2)^{-1/2}\coth\left[\frac{\lambda}{2T}(1-\eta^2x^2)^{1/2}\right]w(x)dx\ , \tag{22a}$$

$$\left[S+\frac{1}{2}\right]-\frac{\lambda\eta^2}{4J}=\frac{1}{2}\int_0^1(1-\eta^2x^2)^{1/2}\coth\left[\frac{\lambda}{2T}(1-\eta^2x^2)^{1/2}\right]w(x)dx\ . \tag{22b}$$

At $\sqrt{1-\eta}\ll T/\lambda\ll 1$ these are as follows:

$$S+\frac{1}{2}=w(1)\frac{T}{\lambda}\left[\frac{1}{2\eta}\ln\left[\frac{1+\eta}{1-\eta}\right]-\ln\left[\frac{2\lambda}{T}\right]\right]+\frac{1}{2}\int_0^1(1-x^2)^{-1/2}w(x)dx+O(T^3)\ , \tag{23a}$$

$$\left[S+\frac{1}{2}\right]-\frac{\lambda\eta^2}{4J}=\frac{1}{2}\int_0^1(1-x^2)^{1/2}w(x)dx+2w(1)\zeta(3)\left[\frac{T}{\lambda}\right]^3+O(T^5)\ . \tag{23b}$$

From these we get

$$1-\eta=\frac{1}{2}\left[\frac{T}{\lambda}\right]^{2\eta}\exp\left[-\frac{2\eta\lambda[m_1+O(T^3)]}{Tw(1)}\right]\ , \tag{24a}$$

$$\lambda=4J\eta^{-2}\left[m_1-2w(1)\zeta(3)\left[\frac{T}{\lambda}\right]^3+O(T^5)\right]\ . \tag{24b}$$

Using $w(1)=2/\pi$ and iteration we have

$$1-\eta=\frac{1}{2}\left[\frac{T}{4Jm_1}\right]^2\exp\left[-\frac{4\pi Jm_0m_1}{T}\right][1+O(T^2)]\ , \tag{25a}$$

$$\lambda=4J\left[m_1-\frac{4}{\pi}\zeta(3)\left[\frac{T}{4Jm_1}\right]^3+O(T^5)\right]\ . \tag{25b}$$

Energy is given by

$$E/N=-2J\left[m_1-\frac{4}{\pi}\zeta(3)\left[\frac{T}{4Jm_1}\right]^3+O(T^5)\right]^2\ . \tag{26}$$

The low-temperature specific heat is $3\zeta(3)T^2/(4\pi J^2m_1^2)$. $1-\eta$ is very small at low temperature. Substituting (25a) and (8c) into (5c) we have asymptotic behavior of $f(\mathbf{r})$ at $(-1)^{\mathbf{r}}=1$, and $g(\mathbf{r})$ at $(-1)^{\mathbf{r}}=-1$ is

$$\frac{T}{\pi^2\eta^2\lambda}\int\int d^2k\frac{\exp(i\mathbf{k}\cdot\mathbf{r})}{k^2+(2\xi)^{-2}} \simeq\frac{2T}{\lambda}\sqrt{\xi/(\pi r)}\exp\left[-\frac{r}{2\xi}\right]\ ,$$

where

$$\xi\equiv[8(\eta^{-2}-1)]^{-1/2} =\frac{\sqrt{2}Jm_1}{T}\exp\left[\frac{2\pi Jm_0m_1}{T}\right][1+O(T^2)]\ . \tag{27a}$$

Correlation function (11) is

$$\langle\mathbf{S}_0\cdot\mathbf{S}_{\mathbf{r}}\rangle\simeq(-1)^{\mathbf{r}}\frac{1}{4\pi}\left[\frac{T}{Jm_1}\right]^2\frac{\xi}{r}\exp\left[-\frac{r}{\xi}\right]\ . \tag{27b}$$

Then ξ is the correlation length in the lattice space unit. This formula for ξ is consistent with Chakravarty, Halperin, and Nelson (CHN).[12] The values of

$$\lim_{T\to 0}T\ln\xi=2\pi Jm_0m_1$$

for $S=\frac{1}{2}$ and 1 are $1.103\,71J$ and $5.446\,56J$. These coincide with AA's numerical results $1.16J$ and $5.46J$.[6] This ξ should be compared with that of ferromagnetic case[4]

$$\xi_F=\sqrt{JS/T}\exp(2\pi JS^2/T)[1+O(T^2)]\ .$$

$S(\mathbf{K})$ in (14) becomes

$$S(\mathbf{K})=\frac{2}{\pi}\exp(4\pi Jm_0m_1/T)[1+O(T^2)]\ . \tag{28}$$

On the contrary, $S(\mathbf{0})$ of square-lattice Heisenberg fermagnet is[4]

$$T(\pi JS)^{-1}\exp(4\pi JS^2/T)\ .$$

Next we consider susceptibility χ in Eq. (13),

$$\chi=\frac{1}{12T}\int_0^1\left[\sinh\left[\frac{\lambda}{2T}(1-\eta^2x^2)^{1/2}\right]\right]^{-2}w(x)dx\ . \tag{29}$$

We should note that this is obtained by operating $-3^{-1}\partial/\partial\lambda$ to the right hand side of (22a). Differentiating the right hand side of (23a) with respect to λ we have

$$\chi=\frac{m_0}{3\lambda}+\frac{w(1)T}{3\lambda^2}+O(T^3) =\frac{m_0}{12Jm_1}+\frac{T}{24\pi J^2m_1^2}+O(T^3)\ . \tag{30}$$

At $T\to 0$ K, χ is

$$(S-0.196\,60)/[12J(S+0.078974)]\ .$$

This value coincides with CHN's spin-wave calculation[12] $(1-0.552/2S)/(12J)$ up to the first term of the $1/S$ expansion. In the classical limit it is $\frac{2}{3}$ times of classical perpendicular susceptibility $1/(8J)$. Then it is expected that

$$m_0/(12Jm_1) \simeq \lim_{T\to 0}\lim_{H\to 0}\chi(T,H) < \lim_{H\to 0}\lim_{T\to 0}\chi(T,H)=\chi_\perp . \quad (31)$$

Point $T=H=0$ is a very singular point of free energy. Moreover, the second term of (30) seems to violate the third law of thermodynamics. Probably the $H=0$ line on the (T,H) plane is a special region and we have $\partial\chi/\partial T=0$ at $H\neq 0$. On the contrary for the 1D $S=\frac{1}{2}$ antiferromagnet[13] we have

$$\lim_{H\to 0}\lim_{T\to 0}\chi(T,H)=\lim_{T\to 0}\lim_{H\to 0}\chi(T,H)$$

and (32)

$$\lim_{T\to 0}\partial\chi/\partial T=0 .$$

AA (Ref 6) have already investigated the subject of this section. But for $1-\eta$ and susceptibility they only gave numerical results of their equation at low temperature. So (27a), (27b), (28), and (30) are our new results.

IV. NUMERICAL RESULTS FOR $S=\frac{1}{2}$ SQUARE LATTICE AND La_2CuO_4

In Table I we show the value of parameter η for the square lattice at $S=\frac{1}{2}$ at a given temperature. This is determined by Eqs. (9a) and (9b). The values of energy per site and magnetic susceptibility are shown. Spin-wave ground-state energy of the 4×4 system is slightly bigger than true value. This is because the theory is based on variational principle. We find that the coincidence is very good at $0\le T\le 0.6J$.

In Fig. 1 we give energy per site by various methods. Lines 1 and 2 are spin-wave results for the 64×64 and 4×4 lattices. Line 3 is the high-temperature expansion up to ninth order,[14]

$$E/N=-0.5J(1.5x+0.75x^2-0.875x^3-1.5625x^4+0.40625x^5+2.705208x^6+0.7137649x^7-4.179204x^8-3.315586x^9),\quad x\equiv J/(2T) . \quad (33)$$

Small circles are results of exact diagonalization for the 4×4 lattice.

In Fig. 2 we give susceptibility per site by various methods. Lines 1 and 2 are our spin-wave results for the 64×64 and 4×4 lattices. Line 3 is the result of high-temperature expansion up to tenth order,[14]

$$\chi=0.25/T(1-2x+2x^2-1.33333x^3+1.083333x^4-1.1833333x^5+0.5097222x^6+0.3218254x^7+0.4073909x^8-1.06728x^9-0.6928188x^{10}) . \quad (34)$$

A similar figure was also given by Okabe *et al.*[15] who calculated χ of the 12×12 and 8×8 lattices using the quantum Monte Carlo method.

Next we analyze the experiments of La_2CuO_4 using our theory. The nearest-neighbor distance is 3.79 Å. From formula (27a) we have

$$\xi=3.79\ \text{Å}\times 0.8186\left[\frac{J}{T}\right]\exp\left[\frac{1.10371J}{T}\right] . \quad (35)$$

The best fit with results of Endoh and co-worker's neutron scattering experiments[3] of ξ^{-1} is obtained at $J=900$ K$=0.0776$ eV$=626$ cm^{-1}. Neutron data and

TABLE I. Spin-wave calculation of η, energy and susceptibility as functions of temperature for the $S=\frac{1}{2}$ 4×4 and 64×64 lattices. The former is compared with an exact diagonalization calculation.

T	η	4×4 $-E/N$ (exact)	χ (exact)
0	0.9929	0.70153(0.70178)	0.0(0.0)
0.1	0.9916	0.69979(0.70145)	0.020388(0.003803)
0.2	0.9871	0.69427(0.69652)	0.042351(0.030216)
0.3	0.9822	0.68696(0.68852)	0.052433(0.047305)
0.4	0.9769	0.67399(0.67472)	0.059879(0.057724)
0.5	0.9710	0.65068(0.64891)	0.067503(0.066822)
0.6	0.9638	0.61376(0.60930)	0.076160(0.075149)

T	$1-\eta$	64×64 $-E/N$	χ
0	$3.E-7$	0.67043	0.0
0.1	$3.45E-5$	0.67016	0.047672
0.2	$7.84E-5$	0.66801	0.051954
0.3	$1.46E-4$	0.66086	0.057199
0.4	$2.78E-4$	0.64345	0.064257
0.5	$6.52E-4$	0.61047	0.073600
0.6	$2.17E-3$	0.55728	0.085808

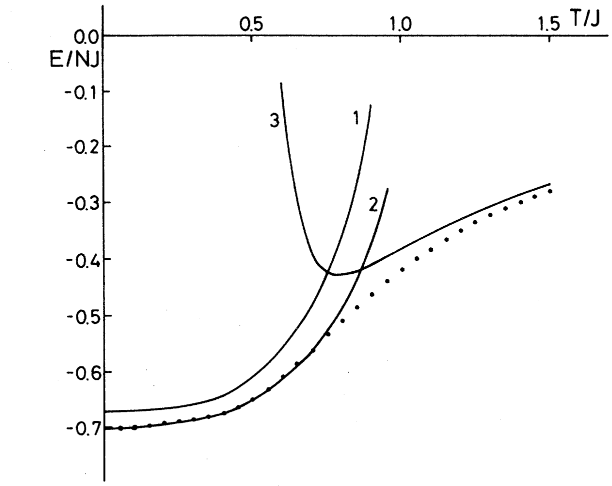

FIG. 1. Energy per site calculated by spin-wave theory for 4×4 and 64×64 lattices. Small circles are results of exact diagonalization of the 4×4 lattice. Agreement is very good in the region $0\le T\le0.7J$. Line 1, spin-wave result of the 64×64 lattice; line 2, spin-wave result of the 4×4 lattice; and line 3, high-temperature expansion.

Eq. (35) are compared in Fig. 3. The formula of spin-wave velocity (21) gives $1.6376J\times3.79$ Å$=0.48$ eV Å. These values are not far from the results of Raman scattering[16] $J=1100\ \mathrm{cm}^{-1}$, $v=0.74$ eV Å. Then we can conclude that the magnetism of La_2CuO_4 is well described by the Hamiltonian (1) and our theory.

At $S=\frac{1}{2}$ Eq. (30) yields

$$\chi=(12J)^{-1}[0.524\,03+0.474\,79(T/J)+O(T^3)]\ .$$

As $\mu_B^2=8.62\times10^{-41}\ \mathrm{erg\,cm}^3$, $k=1.38\times10^{-16}\ \mathrm{erg\,K}^{-1}$,

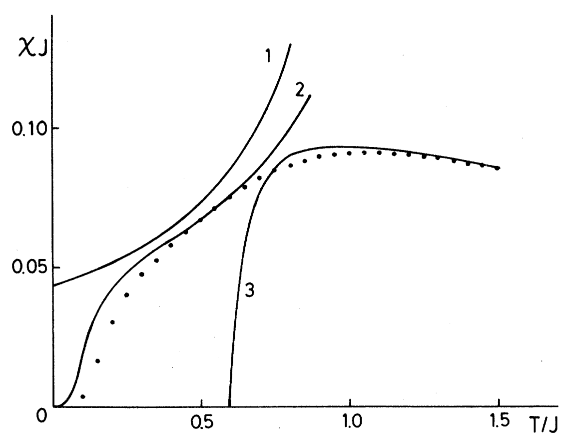

FIG. 2. Susceptibilities per site for uniform magnetic field calculated by spin-wave theory for the $S=\frac{1}{2}$ Heisenberg antiferromagnet on the 4×4 and 64×64 lattices. Small circles are results of exact diagonalization for the 4×4 lattice. Lines 1, 2, and 3 are defined in the caption of Fig. 1.

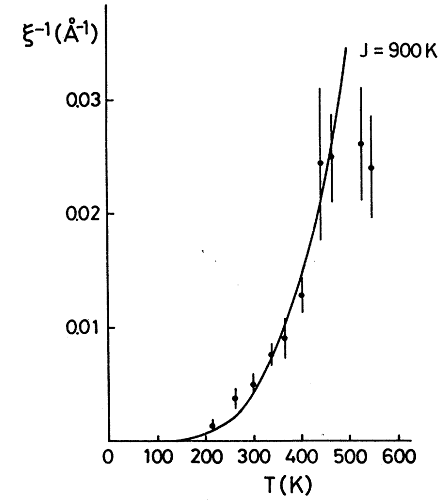

FIG. 3. Inverse of correlation length. Dots with error bars are results of neutron experiment of La_2CuO_4 taken from Ref. 3. The solid line is Eq. (27a) at $J=900$ K.

and $N=1.062\times10^{22}\ \mathrm{cm}^{-3}$, susceptibility of La_2CuO_4 should be

$$\chi=2.457\times10^{-6}\times[0.524\,03+0.474\,79(T/900\ \mathrm{K})+O(T^3)]\ . \qquad (36)$$

V. SUMMARY AND DISCUSSION

At finite temperature and in the limit of infinite system the equations in Sec. II coincide with AA's equations[6] except for the factor of $\frac{3}{2}$ for correlation function (11). So Schwinger boson formulation is not necessary to derive equations in Sec. II for antiferromagnets. Hirsch and Tang got $T=0$ case of our equations using the condition of zero sublattice magnetization. They compared the solution of their equations at $T=0$ with known Monte Carlo results and exact diagonalization. They got very good quantitative coincidence. We obtain a formula of the correlation length which has the same form with CHN.[12] In CHN theory the formula contains two adjustable parameters. But our theory contains only one parameter J and agreement is excellent. According to CHN the formula (35) is the result of one-loop order in renormalization-group theory. In the two-loop–order calculation the preexponential factor may change.

Generally speaking antiferromagnetic spin-wave theory developed in this paper is successful for quantitative calculations of low-temperature properties of square-lattice antiferromagnets. For the two-dimensional square lattice it is known that the ground state has a long-range order at $S\ge1$.[17] In the $S=\frac{1}{2}$ case our theory gives long-range order at the ground state. Presumably this is correct and the value of spontaneous magnetization is not far from the spin-wave value 0.3034.

The case $H\neq0$ of Hamiltonian (1a) may be completely different from the case $H=0$. It is expected that the system has an easy plane if $2JzS>H>0$. The system may have a Kosterlitz-Thouless phase and phase transition. So different spin-wave treatment is necessary for this region.

For one-dimensional systems the ground state of quantum antiferromagnets has no long-range order. So naive application of spin-wave theory formulated in this paper is dangerous. If one applies our theory, the system has finite $1-\eta$ and the two-point function decays exponentially at $T=0$. Elementary excitation has energy gap. These facts qualitatively coincide with Haldane's prediction for integer S systems but strongly contradicts with known algebraic decay of correlation function and gaplessness of the $S=\frac{1}{2}$ system. So further improvement of spin-wave theory is necessary for the 1D antiferromagnet.

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APPENDIX A: CLASSICAL LIMIT

In the Hamiltonian (1a) we try to take the limit of $S\to\infty$ putting $J=J_0S^{-2}$, $H=H_0/S$. Then we have classical Heisenberg Hamiltonian,

$$\mathcal{H}=J_0\sum_{\langle ij\rangle} s_i^xs_j^x+s_i^ys_j^y+s_i^zs_j^z-H_0\sum_i s_i^z,\ \mathbf{s}_i^2=1\ . \tag{A1}$$

In the case $JS\ll T\ll JS^2$ and $H_0=0$, Eqs. (9a) and (9b) become as follows:

$$\frac{zx}{t}=\frac{2}{N}\sum_{\mathbf{k}}{}'\frac{\eta}{1-\eta^2\gamma_{\mathbf{k}}^2}\ , \tag{A2}$$

$$\frac{zx^2}{t}=\frac{2}{N}\sum_{\mathbf{k}}{}'\frac{\eta^2\gamma_{\mathbf{k}}^2}{1-\eta^2\gamma_{\mathbf{k}}^2}\ , \tag{A3}$$

$$t\equiv T/J_0,\quad x\equiv\lambda\eta/(JSz)\ . \tag{A4}$$

Energy per site e is $-J_0x^2z/2$. These equations are equivalent to those for the classical Heisenberg ferromagnet given in Ref. 18. The energy in the 1D and 2D cases are given in the same formulation. The two-point function in (6) is

$$\langle \mathbf{s}_0\cdot\mathbf{s}_{\mathbf{r}}\rangle=\left[\frac{2}{N\beta}\sum_{\mathbf{k}}{}'\frac{\exp(i\mathbf{k}\cdot\mathbf{r})}{1-\eta^2\gamma_{\mathbf{k}}^2}\right]^2 \text{ for } (-1)^{\mathbf{r}}=1\ , \tag{A5}$$

$$\langle \mathbf{s}_0\cdot\mathbf{s}_{\mathbf{r}}\rangle=-\left[\frac{2}{N\beta}\sum_{\mathbf{k}}{}'\frac{\eta\gamma_{\mathbf{k}}\exp(i\mathbf{k}\cdot\mathbf{r})}{1-\eta^2\gamma_{\mathbf{k}}^2}\right]^2 \text{ for } (-1)^{\mathbf{r}}=-1\ , \tag{A6}$$

with $\beta\equiv zx/(\eta t)$. So we find $\langle \mathbf{s}_0\cdot\mathbf{s}_{\mathbf{r}}\rangle_{\mathrm{AF}}=(-1)^{\mathbf{r}}\langle \mathbf{s}_0\cdot\mathbf{s}_{\mathbf{r}}\rangle_{\mathrm{F}}$. Then χ_0 in Ref. 18 corresponds to $S(\mathbf{K})$ in the antiferromagnet. For the square lattice we have

$$E/N=J_0[-2+t+t^2/8+t^3/16+O(t^4)]\ ,$$

$$S(\mathbf{K})\equiv\sum_{\mathbf{r}}(-1)^{\mathbf{r}}\langle \mathbf{s}_0\cdot\mathbf{s}_{\mathbf{r}}\rangle$$

$$=\frac{t^2}{128\pi e^{\pi}}\exp\left[\frac{4\pi}{t}\right]\left[1+\left[\frac{1}{2}-\frac{\pi}{4}\right]t+O(t^2)\right]\ , \tag{A7}$$

$$\xi=\frac{1}{8\sqrt{2}e^{\pi/2}}\exp\left[\frac{2\pi}{t}\right]\left[1-\frac{\pi}{8}t+O(t^2)\right]\ . \tag{A8}$$

Preexponential factor of $S(\mathbf{K})$ and ξ are different from quantum case. In Ref. 18 we compared our spin-wave results of correlation function and those of Monte Carlo results. We obtained very good agreement at low temperature. So we say that our spin-wave equations are in very good agreement with Monte Carlo results. We calculate zero-field susceptibility from (30)

$$\chi\equiv\frac{1}{3T}\sum_{\mathbf{r}}\langle \mathbf{s}_0\cdot\mathbf{s}_{\mathbf{r}}\rangle=\frac{1}{J_0}\left[\frac{1}{12}+\frac{t}{24\pi}+O(t^3)\right]\ . \tag{A9}$$

At $T=0$ and $H_0\neq0$ total energy of (A1) is given by

$$E=-J_02N\cos(2\phi)-NH_0\cos\phi\ , \tag{A10}$$

where ϕ is the angle of the spin relative to the z axis. The minimum is given by $H_0\sin\phi=4J_0\sin(2\phi)$. So we have $\cos\phi=H_0/8J_0$, $\chi(T=0,H_0)=1/8J_0$ at $|H_0|<8J_0$ and $\cos\phi=1$, $\chi=0$ at $|H_0|\geq 8J_0$. We have the following relation for $\chi(T,H_0)$:

$$\frac{1}{12J_0}=\lim_{T\to0}\lim_{H_0\to0}\chi(T,H_0)$$

$$=\frac{2}{3}\lim_{H_0\to0}\lim_{T\to0}\chi(T,H_0)\ . \tag{A11}$$

APPENDIX B: DERIVATION OF (23a) AND (28)

Use the following transformation

$$u\equiv(1-\eta^2x^2)^{1/2}\ . \tag{B1}$$

We put $\eta'\equiv(1-\eta^2)^{1/2}$. Equation (22a) becomes

$$X+\frac{Tw(1)}{\eta\lambda}\int_{\eta'}^{1}\frac{du}{u(1-u^2)^{1/2}}\ , \tag{B2}$$

$$X\equiv\frac{1}{2\eta}\int_{\eta'}^{1}\left[w\left[\frac{(1-u^2)^{1/2}}{\eta}\right]\coth\left[\frac{\lambda u}{2T}\right]-\frac{2Tw(1)}{\lambda u}\right]\frac{du}{(1-u^2)^{1/2}}\ . \tag{B3}$$

As the integrand of (B3) is finite near $u=0$ we have

$$X=O(\eta')+\frac{1}{2}\int_0^1\left[w((1-u^2)^{1/2})\coth\left[\frac{\lambda u}{2T}\right]-\frac{2Tw(1)}{\lambda u}\right]\frac{du}{(1-u^2)^{1/2}}\ .$$

This integral is divided as follows:

$$X=O(\sqrt{1-\eta})+\frac{1}{2}\left[\int_0^1 w((1-u^2)^{1/2})\frac{du}{(1-u^2)^{1/2}}+\int_0^1\left[\coth\frac{\lambda u}{2T}-1\right]\left\{\frac{w((1-u^2)^{1/2})}{(1-u^2)^{1/2}}-w(1)\right\}du\right.$$
$$\left.+w(1)\int_0^1\left[\coth\left[\frac{\lambda u}{2T}\right]-1-\frac{2T}{\lambda u(1-u^2)^{1/2}}\right]du\right].$$

The first integral is

$$\int_0^1 w(x)(1-x^2)^{-1/2}dx\ .$$

The third integral is

$$(2T/\lambda)\ln\{T[1-\exp(-\lambda/T)]/(2\lambda)\}\ .$$

The second integral is $O[(T/\lambda)^3]$ because $\{\ \}$ is $O(u^2)$. Then we get for Eq. (B2)

$$\frac{w(1)T}{\lambda}\left[\frac{1}{2\eta}\ln\left[\frac{1+\eta}{1-\eta}\right]-\ln\left[\frac{2\lambda}{T}\right]\right]+\frac{1}{2}\int_0^1\frac{w(x)}{(1-x^2)^{1/2}}dx+O(T^3)\ .$$

Thus Eq. (23a) is derived.

Using state density function we have for Eq. (14)

$$\frac{1}{4}\int_0^1\frac{1+\eta^2x^2}{1-\eta^2x^2}\left[\coth\left[\frac{\lambda}{2T}(1-\eta^2x^2)^{1/2}\right]\right]^2 w(x)dx-\frac{1}{4}.$$

By transformation (B1) this becomes

$$\frac{1}{4\eta}\int_{\eta'}^1\frac{2-u^2}{u}\left[\coth\frac{\lambda u}{2T}\right]^2\times w\left[\frac{(1-u^2)^{1/2}}{\eta}\right]\frac{du}{(1-u^2)^{1/2}}-\frac{1}{4}\ .$$

Taking the most singular part near $u=0$ we have for Eq. (14)

$$\frac{2w(1)T^2}{\eta\lambda^2}\int_{\eta'}^1\frac{du}{u^3}+O(\ln\eta')=\frac{w(1)T^2}{\eta\lambda^2(1-\eta^2)}+O(\ln\eta')$$
$$=w(1)\exp\left[\frac{4\pi Jm_0m_1}{T}\right][1+O(T^2)]\ ,$$

which equals Eq. (28).

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