Temperature and Spin Dependent Correlation Length of the Quantum Heisenberg Antiferromagnet on the Square Lattice

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The quantum Heisenberg antiferromagnet (HAF) is approached by the *pure-quantum self-consistent harmonic approximation* that reduces it to an effective classical HAF model. The effective exchange enters the classical-like expression for thermal averages as a temperature scale, so that one can obtain in a simple way the quantum spin correlation length from its classical counterpart. For any spin value S the results compare very well with those from experiments, quantum Monte Carlo simulations, and high-T expansion. The adequacy of our theory supports arguments previously raised against the quantitative validity of the mapping of the quantum HAF onto the quantum nonlinear sigma model. [S0031-9007(96)01445-7]

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The square-lattice Heisenberg antiferromagnet (HAF) has attracted much attention in recent years for its connection with the magnetic copper ion planes of high-T_c superconductors and their parent compounds [1]. The model Hamiltonian reads

$$\hat{\mathcal{H}} = \frac{J}{2} \sum_{\mathbf{i},\mathbf{d}} \hat{\mathbf{S}}_{\mathbf{i}} \cdot \hat{\mathbf{S}}_{\mathbf{i}+\mathbf{d}} , \qquad (1)$$

where the index $\mathbf{i} \equiv (i_1, i_2)$ runs over the sites of the square lattice, and $\mathbf{d} \equiv (\pm 1, \pm 1)$ represents the displacements of the 4 nearest neighbors of each site. The quantum spin operators $\hat{\mathbf{S}}_{\mathbf{i}}$ satisfy $|\hat{\mathbf{S}}_{\mathbf{i}}|^2 = S(S+1)$.

The nature of the ground state of this Hamiltonian is a challenging problem, as the existence of an ordered ground state, though rigorously proven for $S \geq 1$ [2], is still not certain for $S = \frac{1}{2}$. Moreover, experimental investigations of several antiferromagnetic compounds with different S show a spin dependence of the thermodynamic quantities which has not yet received a definite theoretical explanation. Most theoretical approaches are based on the seminal ideas of Ref. [3], where the continuum-limit mapping of Eq. (1) into the quantum nonlinear sigma model (QNLσM) is assumed to reproduce its low-wave vector and low-temperature behavior, for any value of the spin.

Using the renormalization group approach the dependence of the critical behavior on the coupling has been studied, on the basis of a correspondence [3,4] that gives the coupling parameter g in terms of the spin stiffness and the spin-wave velocity. From this analysis it appears that real antiferromagnets with Hamiltonian (1) are always in the so-called *renormalized-classical* regime at $T = 0$, so that a classical-like ordered ground state turns out to be appropriate. Unfortunately, the correlation length $\xi(T)$ derived from the QNLσM [5] is not always in agreement with the experimental data and with the high-temperature expansion (HTE) [6] of system (1); this deviation, indeed, increases when S increases, at variance with any semiclassical expansion. Furthermore, it is unclear what is the correct classical parameter $\tilde{S}$ associated with the spin length: S, or $\sqrt{S(S+1)}$, or others like $S + \frac{1}{2}$.

In this Letter we present a different approach, based on the effective Hamiltonian method [7], that maintains the nonuniversal lattice corrections and does not suffer of uncertainty about the expansion parameter.

The mapping onto the QNLσM (when possible) is quite powerful as far as one looks for the existence of fixed points different from the classical one, but the decimation procedure cannot furnish good values of the renormalized parameters, since it treats at the same level both the classical and the quantum part of the fluctuations of high-wave vector modes. Therefore, it is much more appropriate to treat within one-loop (i.e., self-consistent Gaussian) approximation *the purely quantum fluctuations only*, yielding a temperature dependent renormalization of the exchange interaction for a classical-like effective Hamiltonian which contains all the original wave vectors. In this way, one preserves the classical nonlinearities due to the peculiar behavior of classical spin variables moving on a sphere of radius $\tilde{S}$. We then assume an ordered ground state, perturbatively described from the Néel one and determined at the one-loop level.

At variance with previous applications [8] of the effective Hamiltonian method to anisotropic spin systems, the isotropy of the problem prevents us to use the Villain

spin-boson transformation, suitable for easy-plane systems only. Other well-known spin-boson transformations are the Holstein-Primakoff (HP) and the Dyson-Maleev (DM) ones. Both of them break the symmetry of the problem, and at first glance they seem to have inconveniences. Indeed, at finite temperatures the ordered ground state is unstable against low-wave vector thermal fluctuations which, however, have a more and more pronounced classical character; in other words, the symmetry of the isotropic two-dimensional HAF, and hence the vanishing of the order parameter (staggered magnetization), is restored by essentially classical nonlinear excitations.

It is indeed the crucial point of our approach to keep separate the contribution of the *purely quantum* fluctuations from the classical contribution. While the former is evaluated in self-consistent Gaussian approximation, the latter is fully accounted for by means of the effective classical Hamiltonian. The symmetry of the problem can be eventually restored by casting it in the form of a spin Hamiltonian. This permits the use of the HP or the DM spin-boson transformations in a wide range of temperatures.

The procedure leading to the effective Hamiltonian is based on the above-mentioned separation, possible thanks to the path-integral formalism, between classical and purely quantum fluctuations. It is described in Ref. [7], and we named it *pure-quantum self-consistent harmonic approximation* (PQSCHA). In particular, in Refs. [8] it has been used for anisotropic spin systems. The recipe goes through the Weyl symbols [9] for the spin operators; while their explicit form can be determined in the HP framework with a laborious ordering procedure, followed by a resummation [10], at the one-loop level (which we are dealing with) DM is equivalent to HP and turns out to be formally much simpler.

Let us consider a bipartite lattice, consisting of a *positive* and a *negative* sublattice (for a site labeled $\mathbf{i}$ the sublattice sign $(-)^{\mathbf{i}} = \pm 1$ is defined consistently with this terminology). We introduce the DM transformation by writing the spin operators $\hat{S}^z_{\mathbf{i}}$, $\hat{S}^{\pm}_{\mathbf{i}} = \hat{S}^x_{\mathbf{i}} \pm i\hat{S}^y_{\mathbf{i}}$ in terms of boson operators $(\hat{a}^\dagger_{\mathbf{i}}, \hat{a}_{\mathbf{i}})$; for those sitting on positive sites we transform as

$$\hat{S}^+_{\mathbf{i}} = (2S)^{1/2}\hat{a}_{\mathbf{i}}, \qquad \hat{S}^z_{\mathbf{i}} = S - \hat{a}^\dagger_{\mathbf{i}}\hat{a}_{\mathbf{i}};$$
$$\hat{S}^-_{\mathbf{i}} = (2S)^{-1/2}\hat{a}^\dagger_{\mathbf{i}}(2S - \hat{a}^\dagger_{\mathbf{i}}\hat{a}_{\mathbf{i}}), \tag{2}$$

and on the negative sites we use the transformation obtained from the above one replacing $\hat{S}^\mu_{\mathbf{i}} \to -(\hat{S}^\mu)^\dagger$, with $\mu = z, +$, and $-$. Both transformations are canonical (the spin commutation relations are a consequence of $[\hat{a}_{\mathbf{i}}, \hat{a}^\dagger_{\mathbf{i}}] = 1$) and satisfy $|\hat{\mathbf{S}}|^2 = S(S+1)$; furthermore, they are normally ordered in the boson operators $(\hat{a}^\dagger, \hat{a})$, and their replacement in the Hamiltonian (1) gives rise to a normal ordered boson Hamiltonian with quartic interaction. Its normal symbol $\mathcal{H}_{\mathrm{N}}(a^*, a)$ is simply obtained by replacing the Fock operators with commuting holomorphic variables, $(\hat{a}^\dagger, \hat{a}) \to (a^*, a)$. Then, the Weyl symbol [7,9] $\mathcal{H}(a^*, a)$ for $\hat{\mathcal{H}}(\hat{a}^\dagger, \hat{a})$ can be obtained from the normal symbol $\mathcal{H}_{\mathrm{N}}(a^*, a)$ using the relation [9] $\mathcal{H}(a^*, a) = \exp(-\frac{1}{2}\partial_{a^*}\partial_a)\mathcal{H}_{\mathrm{N}}(a^*, a)$.

Using this recipe with the DM spin operators of the positive sublattice (2) it is immediately found that the effective spin length $\tilde{S} \equiv S + \frac{1}{2}$ naturally appears,

$$S^+_{\mathbf{i}} = (2S)^{1/2}a_{\mathbf{i}}, \qquad S^-_{\mathbf{i}} = (2S)^{-1/2}(2\tilde{S} - a^*_{\mathbf{i}}a_{\mathbf{i}})a^*_{\mathbf{i}},$$
$$S^z_{\mathbf{i}} = \tilde{S} - a^*_{\mathbf{i}}a_{\mathbf{i}}; \tag{3}$$

for the negative sublattice the Weyl symbols are obtained by replacing $S^\mu_{\mathbf{i}} \to -(\hat{S}^\mu_{\mathbf{i}})^*$; we have indeed $|\boldsymbol{S}_{\mathbf{i}}|^2 = S^z_{\mathbf{i}}S^z_{\mathbf{i}} + S^+_{\mathbf{i}}S^-_{\mathbf{i}} = \tilde{S}^2$.

Consequently, the Weyl symbol of the Hamiltonian is readily found, but we prefer to express it in terms of the phase-space variables $(p_{\mathbf{i}}, q_{\mathbf{i}})$ that are the Weyl symbols for phase-space operators $(\hat{p}_{\mathbf{i}}, \hat{q}_{\mathbf{i}})$ corresponding to $(\hat{a}^\dagger_{\mathbf{i}}, \hat{a}_{\mathbf{i}})$, such that $[\hat{q}_{\mathbf{i}}, \hat{p}_{\mathbf{i}}] = i\tilde{S}^{-1}$. Eventually, the Weyl symbol of the boson Hamiltonian becomes

$$\mathcal{H} = -\frac{J\tilde{S}^2}{2}\sum_{\mathbf{i},\mathbf{d}}\Big[(1 - z^2_{\mathbf{i}})(1 - z^2_{\mathbf{i}+\mathbf{d}}) + \left(1 - \frac{z^2_{\mathbf{i}} + z^2_{\mathbf{i}+\mathbf{d}}}{4}\right)(q_{\mathbf{i}}q_{\mathbf{i}+\mathbf{d}} - p_{\mathbf{i}}p_{\mathbf{i}+\mathbf{d}}) + i(-)^{\mathbf{i}}\frac{z^2_{\mathbf{i}} - z^2_{\mathbf{i}+\mathbf{d}}}{4}(q_{\mathbf{i}}p_{\mathbf{i}+\mathbf{d}} + p_{\mathbf{i}}q_{\mathbf{i}+\mathbf{d}})\Big], \tag{4}$$

where $z^2_{\mathbf{i}} \equiv (q^2_{\mathbf{i}} + p^2_{\mathbf{i}})/2$. Its minimum configuration is the Néel one, given by $\{p_{\mathbf{i},0} = 0, q_{\mathbf{i},0} = 0\}$.

The corresponding effective Hamiltonian is easily found to be

$$\mathcal{H}_{\mathrm{eff}} = -\frac{J\tilde{S}^2}{2}\sum_{\mathbf{i},\mathbf{d}}\Big[1 - \theta^2(z^2_{\mathbf{i}} + z^2_{\mathbf{i}+\mathbf{d}}) + z^2_{\mathbf{i}}z^2_{\mathbf{i}+\mathbf{d}} + \left(\theta^2 - \frac{z^2_{\mathbf{i}} + z^2_{\mathbf{i}+\mathbf{d}}}{4}\right)(q_{\mathbf{i}}q_{\mathbf{i}+\mathbf{d}} - p_{\mathbf{i}}p_{\mathbf{i}+\mathbf{d}}) + i(-)^{\mathbf{i}}\frac{z^2_{\mathbf{i}} - z^2_{\mathbf{i}+\mathbf{d}}}{4}(q_{\mathbf{i}}p_{\mathbf{i}+\mathbf{d}} + p_{\mathbf{i}}q_{\mathbf{i}+\mathbf{d}})\Big] + T\sum_{\mathbf{k}}\ln\frac{\sinh f_k}{f_k} - NJ\tilde{S}^2\frac{\mathcal{D}^2}{2}. \tag{5}$$

The renormalization parameter $\theta^2 \equiv 1 - \mathcal{D}/2$ represents the effect of pure-quantum fluctuations,

$$\mathcal{D} = \frac{1}{\tilde{S}N}\sum_{\mathbf{k}}(1 - \gamma^2_{\mathbf{k}})^{1/2}\left(\coth f_{\mathbf{k}} - \frac{1}{f_{\mathbf{k}}}\right), \tag{6}$$

where $\gamma_{\mathbf{k}} = (\cos k_x + \cos k_y)/2$, and depends on the frequency spectrum through $f_{\mathbf{k}} = \omega_{\mathbf{k}}/(2\tilde{S}T)$. We recall [7] that in order to manage with the PQSCHA in a system with many degrees of freedom we must also apply the so-called "low-coupling approximation." The low and

intermediate temperature calculation of the pure-quantum renormalization parameters deserves a more accurate approach, since we are dealing with a system with strong classical anharmonicity. In this temperature range we have to take the fully renormalized (i.e., including the classical contribution) frequency spectrum. At higher temperatures, $\mathcal{D}$ decreases and is less dependent on the frequency spectrum as $f_{\mathbf{k}}$ tends to vanish. Therefore we have the self-consistent definition

$$f_{\mathbf{k}} = \frac{2e(t)}{\tilde{S}t}(1 - \gamma_{\mathbf{k}}^2)^{1/2}, \tag{7}$$

where $t \equiv T/(J\tilde{S}^2)$ is the reduced temperature; the frequency renormalization parameter $e(t)$ is $e(t) = \theta^2(t)$ for $t \to \infty$, while at low temperature $e(t) = \kappa^2(t) \equiv 1 - (\mathcal{D} + \mathcal{D}_{\mathrm{cl}})/2 = \theta^2 - \mathcal{D}_{\mathrm{cl}}/2$, with

$$\mathcal{D}_{\mathrm{cl}} = \frac{1}{\tilde{S}N}\sum_{\mathbf{k}}(1 - \gamma_{\mathbf{k}}^2)^{1/2}\frac{1}{f_{\mathbf{k}}} = \frac{t}{2\kappa^2}. \tag{8}$$

For increasing temperature, due to the lack of long-range order, only spin waves with wavelength $\lambda \lesssim 2\xi$ (ξ is the spin correlation length) survive in the system. Neglecting this, the well-known instability of the frequency renormalization κ^2 is found for $t = \theta^4$. This unphysical feature can be washed out by inserting a correlative cutoff (in the antiferromagnetic Brillouin zone) in calculating $\mathcal{D}_{\mathrm{cl}}$, thus smoothly connecting with the high-temperature regime.

By rescaling the classical phase-space variables as $(p_{\mathbf{i}}, q_{\mathbf{i}}) \to (\theta\, p_{\mathbf{i}}, \theta\, q_{\mathbf{i}})$ the effective Hamiltonian can be cast in the same form of Eq. (4), multiplied by the factor θ^4, plus uniform terms. Then, using the classical counterpart of the DM transformation (2) for spins of length $\theta^2\tilde{S}$, we transform to classical spin variables $\theta^2\tilde{S}\boldsymbol{s}$, such that $|\boldsymbol{s}| = 1$. We eventually have

$$\mathcal{H}_{\mathrm{eff}} = -\frac{J\tilde{S}^2\theta^4}{2}\sum_{\mathbf{i},\mathbf{d}}\boldsymbol{s}_{\mathbf{i}} \cdot \boldsymbol{s}_{\mathbf{i}+\mathbf{d}} + NJ\tilde{S}^2\mathcal{G}(t). \tag{9}$$

The term $\mathcal{G}(t) = tN^{-1}\sum_{\mathbf{k}} \ln[\sinh f_k/(\theta^2 f_k)] - 2\theta^2\mathcal{D}$ is uniform and does not play any role in calculating thermal averages. In this Hamiltonian the effects of quantum fluctuations are given by

(i) the log term that transforms the spin-wave contribution to the free energy from classical to quantum;

(ii) the renormalized exchange integral $J \to J_{\mathrm{eff}} = \theta^4 J$ that depends on the temperature t and on the spin quantum number S;

(iii) the appearance of the factor $\tilde{S} = S + \frac{1}{2}$ as the classical spin length for the semiclassical renormalization approach; this is a direct consequence of the PQSCHA recipe, without any empirical observation.

At lowest temperature $\mathcal{H}_{\mathrm{eff}}$ reproduces all the results of the self-consistent harmonic approximation (SCHA), giving the one-loop renormalization effect as long as the self-consistent Eq. (8) for κ^2 admits a positive solution. At $t = 0$, $\kappa^2 = \theta^2$ gives the one-loop quantum correction to the spin stiffness, and decreases with decreasing $\tilde{S}$; the instability value $\theta^2 = 0$ is not reached using physical values of S. At high temperature $\mathcal{H}_{\mathrm{eff}}$ approaches the classical situation. At intermediate temperature there is an interval, whose width is larger the smaller is S, where nonlinear quantum effects (due to higher order terms in the coupling) are significant; this fact could be interpreted as the presence of the so-called *quantum-critical* regime (QCR) [3,4].

The parameter $\theta^4(S,t)$ gives the temperature dependent effects of the quantum fluctuations on the intensity of the exchange for different values of the spin. The PQSCHA expression of the spin-spin correlation function in terms of a classical-like average with the effective Hamiltonian is $(-)^{\mathbf{r}}\langle\hat{\mathbf{S}}_{\mathbf{i}} \cdot \hat{\mathbf{S}}_{\mathbf{i}+\mathbf{r}}\rangle = (-)^{\mathbf{r}}\tilde{S}^2\theta_{\mathbf{r}}^4\langle\boldsymbol{s}_{\mathbf{i}} \cdot \boldsymbol{s}_{\mathbf{i}+\mathbf{r}}\rangle_{\mathrm{eff}}$, and $\langle\boldsymbol{s}_{\mathbf{i}} \cdot \boldsymbol{s}_{\mathbf{i}+\mathbf{r}}\rangle_{\mathrm{eff}}$ appears to be equal to the classical-limit average, but at the temperature $t_{\mathrm{cl}} = t/\theta^4(S,t)$.

The parameter $\theta_{\mathbf{r}}^4$ is related to the fluctuation of spins at a distance $\mathbf{r}$, and it tends to a constant as $r = |\mathbf{r}|$ increases. This means that the temperature behavior of the quantum correlation length $\xi(t)$ is connected with its classical counterpart $\xi_{\mathrm{cl}}(t)$ by the equality

$$\xi(t) = \xi_{\mathrm{cl}}(t_{\mathrm{cl}}), \qquad t_{\mathrm{cl}} = t/\theta^4(t), \tag{10}$$

i.e., $\xi(t)$ can be obtained for any spin length starting from the classical $\xi_{\mathrm{cl}}(t)$. Values for the latter in the range $1 \lesssim \xi \lesssim 8$ have been obtained by Monte Carlo simulation [11] and by HTE [6].

We ourselves have performed some Monte Carlo simulations in order to extend this data range: using a 256×256 lattice the classical values we have determined are $\xi(0.70) = 7.8 \pm 0.1$, $\xi(0.65) = 13.2 \pm 0.3$, $\xi(0.60) = 27.3 \pm 0.5$, and $\xi(0.57) = 52.0 \pm 1.0$. All the available classical data sets agree with each other,

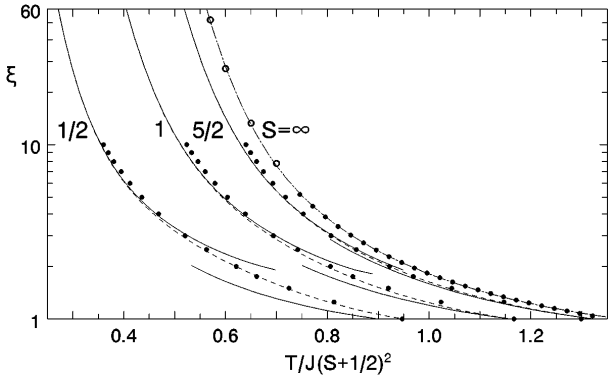

FIG. 1. Correlation length $\xi(t)$ vs reduced temperature $t = T/J\tilde{S}^2$, for spin $S = \infty$ (classical), $\frac{5}{2}$, 1, and $\frac{1}{2}$. The continuous lines are the low- and high-temperature results of our theory, and the dashed line represents the application of the cutoff condition for long-wavelength spin waves. The filled circles are the classical and quantum results from high-temperature expansion [6]; the open circles are our new Monte Carlo simulation results. The dash-dotted line is a fit to the classical data.

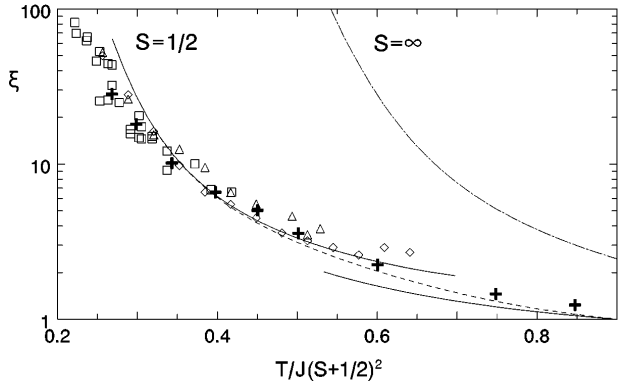

FIG. 2. Correlation length $\xi(t)$ for spin $S = \frac{1}{2}$. Squares: experimental data [12,13] for $Sr_2CuO_2Cl_2$; triangles and diamonds: data for La_2CuO_4 from neutron scattering [15] and from ^{63}Cu NQR relaxation [14] experiments, respectively; crosses: quantum Monte Carlo results [16]. Lines as in Fig. 1.

so we have used the data of Ref. [6] and ours, and fitted them by a (polynomial $\times$ exponential) curve in the range $1 \lesssim \xi \lesssim 50$.

The quantum counterparts of this classical curve, obtained by renormalization of the temperature scale for the spin length values $S = \frac{1}{2}$, 1, and $\frac{5}{2}$, are plotted in Fig. 1 together with the HTE results [6].

In Fig. 2 we report our result for $\xi(t)$ at spin $S = \frac{1}{2}$ together with experimental data for $Sr_2CuO_2Cl_2$ [12,13], for La_2CuO_4 [14,15], and with quantum Monte Carlo results [16]. The region where nonlinear quantum effects are relevant ranges from $t \gtrsim 0.5$ to $t \lesssim 0.75$, which agrees with the range of QCR predicted in Ref. [4].

In Fig. 3 we compare our curve at spin $S = 1$ with experimental data for La_2NiO_4 [17] and for K_2NiF_4 [13]. For this spin value, the quantum effects are less relevant and quantum nonlinear effects are reduced within a shorter interval.

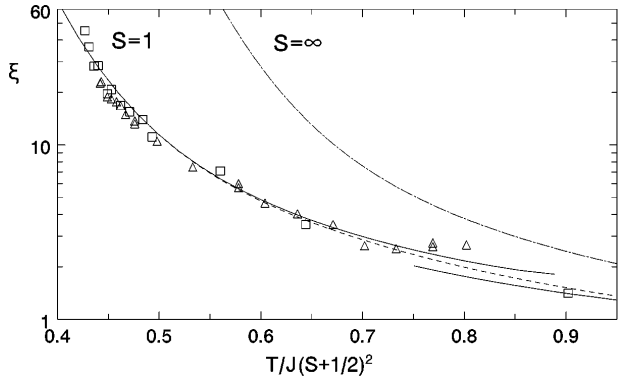

FIG. 3. Correlation length for spin $S = 1$. Triangles: experimental data [17] for La_2NiO_4; squares: experimental data [13] for K_2NiF_4. Lines as in Fig. 1.

Our results appear to explain all the experimental data [12–15,17] for different values of S without any fitting parameters. They also agree with the HTE results of Ref. [6], thus confirming the inadequacy of the approach by mapping the quantum HAF onto the QNLσM. Most importantly, however, we can move to much lower temperatures where significant experimental data are available. On the other hand, the good agreement we still find for $S = \frac{1}{2}$ is an indirect proof that the ground state is ordered and that for nearest-neighbor interaction the critical value of S is smaller than any physical spin value. Finally, we approach with continuity the high-T region, also owing to the presence of the effective spin length $\tilde{S}$ unambiguously determined by the theory itself.

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