Thermodynamics of the quantum easy-plane antiferromagnet on the triangular lattice

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The classical XXZ triangular-lattice antiferromagnet shows both an Ising and a Berezinskii-Kosterlitz-Thouless transition, related to the chirality and the in-plane spin components, respectively. In this paper the quantum effects on the thermodynamic quantities are evaluated by means of the pure-quantum self-consistent harmonic approximation, which allows one to deal with any spin value through classical Monte Carlo simulations. We present data for the internal energy, the specific heat, the static spin correlation functions, and the in-plane correlation length for different values of the spin and of the exchange anisotropy. The quantum transition temperatures turn out to be smaller the smaller the spin, and agree with the few available theoretical and numerical estimates. [S0163-1829(99)04234-4]

A renewed interest has recently focused on triangular antiferromagnets (TAF's).[1] Indeed, they turned out to describe the magnetic behavior of several real compounds such as, for example, the stacked antiferromagnet $NaTiO_2$,[2] the organic superconductors of the family $\kappa-(BEDT-TTF)_2X$,[3] and the K/Si(111):B interface.[4]

In this paper we investigate the thermodynamic properties of the quantum XXZ Heisenberg antiferromagnet on the triangular lattice, defined by the following Hamiltonian:

$$\hat{\mathcal{H}}=\frac{J}{2}\sum_{\mathbf{i},\mathbf{d}}\left(\hat{S}^x_{\mathbf{i}}\hat{S}^x_{\mathbf{i}+\mathbf{d}}+\hat{S}^y_{\mathbf{i}}\hat{S}^y_{\mathbf{i}+\mathbf{d}}+\lambda\,\hat{S}^z_{\mathbf{i}}\hat{S}^z_{\mathbf{i}+\mathbf{d}}\right), \tag{1}$$

where J is the positive (antiferromagnetic) exchange constant, and $(\hat{S}^x_{\mathbf{i}},\hat{S}^y_{\mathbf{i}},\hat{S}^z_{\mathbf{i}})$ are the spin operators sitting on the sites $\mathbf{i}$ of a triangular lattice. They satisfy $\mathcal{SU}(2)$ commutation relations and belong to the spin-S representation $|\hat{\mathbf{S}}^2_{\mathbf{i}}|=S(S+1)$. The interaction is restricted to nearest neighbors and $\mathbf{d}$ runs over their relative displacements. The planar character of the system is due to the presence of the anisotropy $\lambda\in[0,1)$, energetically favoring configurations with the spins lying in the xy plane (easy plane). For $\lambda=0$ the spin components on the z axis do not appear in the Hamiltonian and the model is known as $XX0$ or quantum XY.

The minimum energy configuration of the classical counterpart of the Hamiltonian (1) for every value of $\lambda\in[0,1]$ consists of coplanar spins forming $\pm 2\pi/3$ angles between nearest neighbors and this leads to a $\sqrt{3}\times\sqrt{3}$ periodic Néel state. In contrast to the isotropic case, where the plane in which the $2\pi/3$ structure lies can take any direction in the spin space, in the XXZ model such structure must take place in the easy plane. As a result, in the planar TAF the frustration causes an additional discrete twofold degeneracy of the classical ground state, which is due to the chirality (or helicity), defined as the sign of rotation of the spins along the sides of each elementary triangle. The resulting degeneracy corresponds to the group $SO(2)\times Z_2$. As the Mermin-Wagner theorem only states that the sublattice magnetization must vanish at any nonzero temperature, long-range order can occur as far as the chirality is concerned, and an Ising-like phase transition is indeed observed,[5] in addition to the usual Berezinskii-Kosterlitz-Thouless (BKT) critical behavior associated with the rotation symmetry in the xy plane.

In the quantum case the situation is far less clear. In fact, unlike the antiferromagnet on the square lattice where there is a general consensus about the ordered nature of the ground state even for $S=1/2$, in the frustrated cases the lack of exact analytical results is accompanied by difficulties in applying stochastic numerical methods, as their reliability is strongly limited by the well-known sign problem. Indeed only very recently a systematic size scaling of the order parameter and of the spin gap has been performed using a new quantum Monte Carlo technique,[6] confirming the existence of Néel long-range order in the ground state as also suggested by the symmetry properties of the first excited states, evidenced by Bernu *et al.*[7]

An even less clear situation is that concerning the finite-temperature behavior. In fact an early numerical work,[8] limited to lattice sizes up to 27 sites, indicated for the $S=1/2$ $XX0$ model a phase diagram similar to the classical one, in contrast with the high-temperature expansion produced by Fujiki and Betts[9] where no evidence for a phase transition was found. For the XXZ Hamiltonian, Momoi and Suzuki,[10] applying an effective field theory, conjectured that the chiral phase transition should persist for every value of

$\lambda \in [0,1)$, as in the classical case, and were able to estimate the transition temperature for $\lambda = 0$, obtaining a value very close to that found in Ref. 8. Recently Suzuki and Matsubara,[11] using a quantum transfer Monte Carlo method to study clusters up to 24 sites, have claimed instead the absence of the chiral order at any finite temperature for $\lambda \geqslant 0.6$.

In this context where, at least up to now, quantum numerical methods cannot give satisfactory answers, the *pure-quantum self-consistent harmonic approximation*[12] (PQSCHA) can provide an effective instrument to investigate the thermodynamics of quantum spin systems, as far as their ground state is ordered. The method is based on the path-integral formulation of quantum statistical mechanics, and has been successfully applied recently to a variety of unfrustrated spin models, both one[13] and two dimensional.[14,15]

By the PQSCHA the evaluation of thermal averages in the quantum model can be reduced to the calculation of classical-like averages over a Boltzmann distribution defined by an effective Hamiltonian, which contains the contribution of the pure-quantum part of the fluctuations (approximated within a self-consistent harmonic scheme) in its renormalized interaction parameters, which are temperature and spin dependent. As a consequence one can get accurate results on the quantum spin system using classical computational methods, like the transfer matrix in the one-dimensional case and classical Monte Carlo (MC) simulations in the two-dimensional one.

The first step of the derivation of the effective Hamiltonian for the easy-plane TAF is to apply the unitary transformation which defines a spatially varying coordinate system pointing along the local Néel direction, namely,

$$\hat{\mathcal{U}} = \exp\left(\frac{2\pi}{3} i \sum_{\mathbf{i} \in B} \hat{S}^z_{\mathbf{i}} - \frac{2\pi}{3} i \sum_{\mathbf{i} \in C} \hat{S}^z_{\mathbf{i}} \right), \tag{2}$$

where B and C label two of the three sublattices. Unlike in the bipartite lattices where the corresponding transformation maps the antiferromagnet into a model with an in-plane ferromagnetic exchange and an antiferromagnetic coupling along the z axis, thus allowing the demonstration of the Lieb-Mattis theorem[16] and computability with standard quantum Monte Carlo methods, in the triangular case the transformed Hamiltonian shows an extra currentlike term[17] which contains the effects of the frustration, whose form is quite similar to the chiral order parameter,[5] i.e., the physical quantity undergoing the order-disorder phase transition present in the classical model for every value of $\lambda < 1$.

From now on the derivation follows the same lines already described in Refs. 13 and 15. A point worth being recalled is the use of the Villain transformation[18] in order to represent the spin operators in terms of canonically conjugated variables, which is a necessary step in the derivation of the effective Hamiltonian. As is well known, this spin-boson transformation preserves the commutation rules but neglects the so-called kinematic interaction due to the limited spectrum of S^z, thus giving a better description when the system has a good easy-plane character and the spin states with large fluctuations of S^z are less relevant to the thermodynamics. In the square lattice case[15] such an approximation scheme turns out to be reliable up to some value $\lambda_M < 1$ of λ ($\lambda_M = 0.58$ in the extreme quantum case $S = 1/2$), when the mapping with the Villain transformation breaks down and a different spin-boson transformation is needed. However, it provides accurate results for the critical temperatures even for $\lambda = 0.5$; a similar behavior is also found in the case of the quantum TAF. Finally we remind that Weyl ordering, which is inherent to the PQSCHA, naturally leads to define an effective classical spin length as $\tilde{S} = S + 1/2$ and thus to set the natural energy scale $\epsilon = J\tilde{S}^2$. Therefore in the following we use the reduced temperature $t = k_B T / J\tilde{S}^2$.

In the case of the easy-plane TAF the effective Hamiltonian has the form $\mathcal{H}_{\text{eff}} = \overline{\mathcal{H}} + G(t)$, where $G(t)$ is an additive uniform term, formally identical to that obtained for the square lattice, which is unessential in the calculation of the thermal averages, while

$$\overline{\mathcal{H}} = \frac{\epsilon}{2} j_{\text{eff}} \sum_{\mathbf{i},\mathbf{d}} \left(s^x_{\mathbf{i}} s^x_{\mathbf{i}+\mathbf{d}} + s^y_{\mathbf{i}} s^y_{\mathbf{i}+\mathbf{d}} + \lambda_{\text{eff}} s^z_{\mathbf{i}} s^z_{\mathbf{i}+\mathbf{d}} \right), \tag{3}$$

where $(s^x_{\mathbf{i}}, s^y_{\mathbf{i}}, s^z_{\mathbf{i}})$ are unit vectors, i.e., classical spins. Within the PQSCHA quantum effects are embodied in the temperature and spin dependence of the renormalized interaction parameters

$$j_{\text{eff}}(t,S,\lambda) = \left(1 - \frac{1}{2} D_\perp \right)^2 e^{-\mathcal{D}_\parallel / 2}, \tag{4}$$

$$\lambda_{\text{eff}}(t,S,\lambda) = \lambda \left(1 - \frac{1}{2} D_\perp \right)^{-1} e^{\mathcal{D}_\parallel / 2}, \tag{5}$$

with

$$D_\perp = (2\tilde{S}N)^{-1} \sum_{\mathbf{k}} \frac{b_{\mathbf{k}}}{a_{\mathbf{k}}} \mathcal{L}(f_{\mathbf{k}}), \tag{6}$$

$$\mathcal{D}_\parallel = (\tilde{S}N)^{-1} \sum_{\mathbf{k}} (1 - \gamma_{\mathbf{k}}) \frac{a_{\mathbf{k}}}{b_{\mathbf{k}}} \mathcal{L}(f_{\mathbf{k}}), \tag{7}$$

where

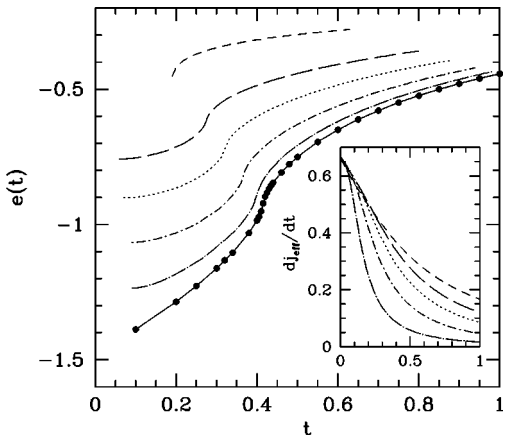

FIG. 1. Temperature and spin dependence of the internal energy per spin for $\lambda = 0$ and (from the top curve) $S = 1/2$, 1, 3/2, 5/2, 5, and ∞. Solid circles are classical MC data. In the inset the derivative of the effective exchange constant is plotted vs t with the same convention on the lines.

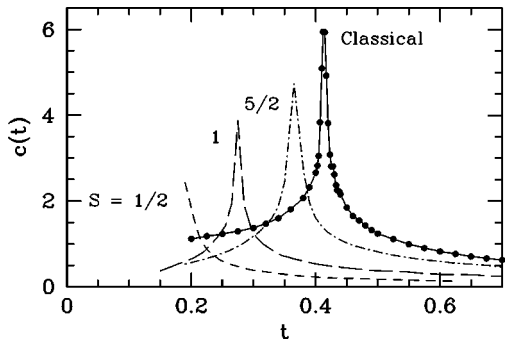

FIG. 2. Temperature and spin dependence of the specific heat in the $XX0$ model. Circles are the classical MC data obtained from the mean-squared fluctuations of the energy while the solid line is the numerical derivative of the energy curve.

$$a_{\mathbf{k}}^2=\frac{z}{2}(1-\tfrac{1}{2}D_\perp)\mathrm{e}^{-\mathcal{D}_\parallel/2}(1+2\lambda_{\mathrm{eff}}\,\gamma_{\mathbf{k}}), \tag{8}$$

$$b_{\mathbf{k}}^2=\frac{z}{2}j_{\mathrm{eff}}(1-\gamma_{\mathbf{k}}), \tag{9}$$

$f_{\mathbf{k}}=a_{\mathbf{k}}b_{\mathbf{k}}/(2\tilde{S}t)$, $\mathcal{L}(x)=\coth x-x^{-1}$ is the Langevin function, $\gamma_{\mathbf{k}}=z^{-1}\Sigma_{\mathbf{d}}\cos(\mathbf{k}\cdot\mathbf{d})$, $z=6$ is the coordination number of the lattice, and $\mathbf{k}$ is a wave vector varying in the first Brillouin zone. $D_\perp(S,\lambda,t)$ and $\mathcal{D}_\parallel(S,\lambda,t)$ represent the pure-quantum square fluctuations of the out-of-plane and in-plane components of the spins, respectively. They are decreasing functions of temperature and spin, vanishing for $t\to\infty$ and $S\to\infty$, i.e., when the quantum part of the fluctuations is negligible with respect to the classical one.

From the above equations, we can infer that the PQSCHA approach is valid under the condition that second-order terms in $(D_\perp)^2$ can be neglected. One can take the criterion that the renormalization effects of quantum fluctuations must not reduce much more than, say, 50% the effective exchange integral. Such a strong renormalization only occurs for $S=1/2$ and $t\lesssim 0.2$, while for higher spin values the PQSCHA is reliable at any temperature.

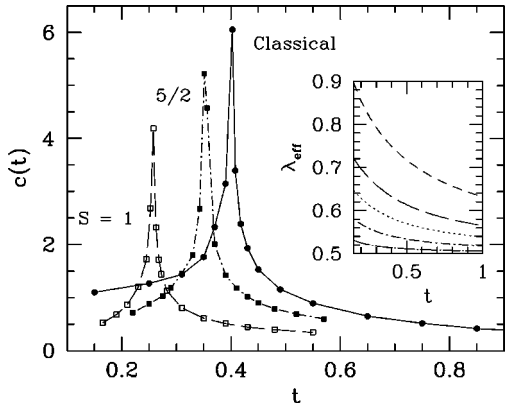

FIG. 3. Specific heat for the XXZ model with $\lambda=0.5$ as a function of the temperature. Open squares, $S=1$; solid squares, $S=5/2$; solid circles, $S=\infty$ (classical). The inset displays the temperature dependence of λ_{eff} for $\lambda=0.5$ and $S=1/2$, 1, 3/2, 5/2, 5, with the same convention on the lines of Fig. 1.

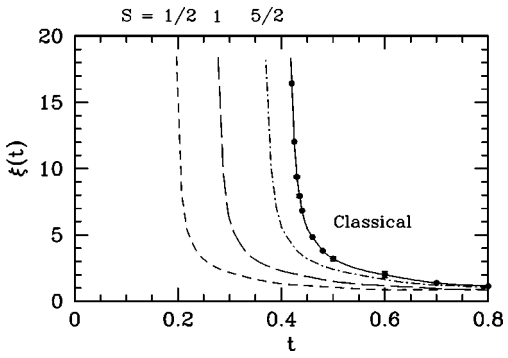

FIG. 4. Temperature and spin dependence of the magnetic correlation length in the $XX0$ model. Circles and the solid line are classical MC data.

In the $XX0$ model, $\lambda_{\mathrm{eff}}=\lambda=0$ and all the information about the quantum system is hence contained in the renormalization of the energy scale. In this case the critical properties of the quantum system at a temperature t are essentially those of its classical counterpart at the effective temperature $t_{\mathrm{eff}}=t/j_{\mathrm{eff}}(t,S)$, and we have used the results of classical Monte Carlo simulations recently obtained for lattice sizes up to $N=120\times 120^5$ to calculate the corresponding quantum observables.

Indicating the classical averages with the effective Hamiltonian as $\langle\cdots\rangle_{\mathrm{eff}}$, the internal energy per spin can be calculated as

$$e(t,S,\lambda)=\frac{\langle\hat{\mathcal{H}}\rangle}{N\epsilon}=\langle\overline{\mathcal{H}}\rangle_{\mathrm{eff}}+\frac{z}{2}\lambda\,\mathcal{D}_\perp\,, \tag{10}$$

where $\mathcal{D}_\perp$ can be expressed as $D_\perp$, Eq. (6), with an extra factor $\gamma_{\mathbf{k}}$ in the summand. For $\lambda=0$ the above equation reduces to

$$e(t,S)=j_{\mathrm{eff}}(t)\,e_{\mathrm{cl}}(t_{\mathrm{eff}}), \tag{11}$$

$e_{\mathrm{cl}}(t)$ being the internal energy per spin of the corresponding classical system. In Fig. 1 $e(t,S)$ is plotted for various values of the spin in the range of temperatures where the PQSCHA is expected to give reliable results.

The energy curves flatten and increase with decreasing S due to the increased quantum fluctuations. As said before the

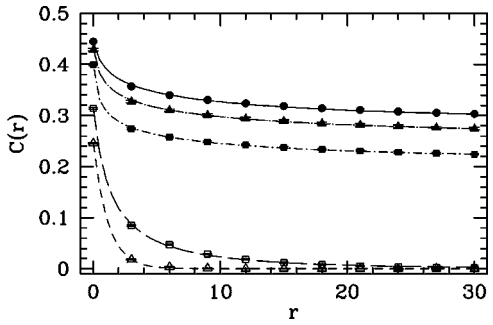

FIG. 5. In-plane normalized spin-spin correlation function for the XXZ model with $\lambda=0.5$, $t=0.27$, and $S=1/2$ (open triangles), $S=1$ (open squares), $S=5/2$ (solid squares), $S=5$ (solid triangles), and $S=\infty$ (solid circles). Lines are best fits against the expected high-temperature ($S=1/2$ and $S=1$) and low-temperature ($S\geqslant 5/2$) asymptotical decay of the correlation functions.

TABLE I. Chiral and BKT critical temperatures for $\lambda=0$ and $\lambda=0.5$ and for some values of the spin length S. The classical values are taken from Ref. 5. The reported errors only represent the statistical uncertainty of the MC data.

S	1/2	1	3/2	5/2	5	∞
$t_c(S,0)$	0.193(2)	0.273(3)	0.319(3)	0.364(4)	0.396(5)	0.412(5)
$t_{\mathrm{BKT}}(S,0)$	0.1875(5)	0.265(1)	0.310(1)	0.352(1)	0.386(1)	0.402(2)
$t_c(S,0.5)$	0.185(4)	0.267(4)	0.312(5)	0.355(5)	0.385(5)	0.400(5)
$t_{\mathrm{BKT}}(S,0.5)$	0.180(1)	0.260(1)	0.304(2)	0.346(2)	0.376(2)	0.391(2)

$S=1/2$ curve is reported only in the valid temperature range. As a matter of fact the extrapolation to lowest temperatures gives the self-consistent spin-wave ground-state energy. The difference from the most refined estimates[17] can be mainly attributed to $1/S^2$ constant contributions coming from the Villain transformation[19] and to the use of the low coupling approximation (LCA).[12] This term is not significant for $S \geqslant 1$.

Consistently with this picture the finite-size ($N=120\times120$) peak of the specific heat (Fig. 2), obtained by numerical derivation of the internal energy, moves towards lower temperatures and decreases in height as S decreases. However, since the quantum renormalizations are essentially size independent, classical scaling with size[5] is conserved and a logarithmic divergence of the specific heat, connected with the Ising-like chirality phase transition, is therefore expected in the thermodynamic limit. By direct derivation of Eq. (11) it is easily seen that, in order for the quantum specific heat to vanish in the zero-temperature limit, within our approximation we must have $d\,j_{\mathrm{eff}}/dt \to |e_{\mathrm{cl}}(0)|^{-1}$ as $t\to0$, a condition which is fulfilled for every value of S, as can be verified analytically from the explicit expressions of the renormalization parameters; this is shown in the inset of Fig. 1.

For $\lambda\neq0$ quantum effects cannot be embodied in a mere redefinition of the energy scale. In fact they are also contained in the temperature and spin-dependent renormalizations of the easy-plane anisotropy $\lambda_{\mathrm{eff}}(t,S,\lambda)$ (sketched in the inset of Fig. 3). We have therefore calculated the physical properties of the XXZ model by performing a new set of classical Monte Carlo simulations using the same algorithm and lattice sizes of Ref. 5. In particular, for $\lambda=0.5$ the specific heat behavior of the quantum XXZ model, shown in Fig. 3 for $S\geqslant1$, turns out to be quite similar to that observed for $\lambda=0$.

Most papers in the literature are mainly concerned with the chiral order-disorder transition, while the XXZ TAF also supports another kind of phase transition. In fact, the classical system[5] displays as well a BKT critical behavior. For this reason we have calculated the magnetic correlation length which governs the decay of the in-plane correlation functions in the high-temperature phase, whose expression within the PQSCHA reads

$$C(\mathbf{r})=\tilde{S}^{-2}\langle \hat{S}^x_{\mathbf{i}}\hat{S}^x_{\mathbf{i}+\mathbf{r}}+\hat{S}^y_{\mathbf{i}}\hat{S}^y_{\mathbf{i}+\mathbf{r}}\rangle = G(\mathbf{r})\,\langle s^x_{\mathbf{i}}s^x_{\mathbf{i}+\mathbf{r}}+s^y_{\mathbf{i}}s^y_{\mathbf{i}+\mathbf{r}}\rangle_{\mathrm{eff}}, \tag{12}$$

where $\mathbf{i}$ and $\mathbf{i}+\mathbf{r}$ belong to the same sublattice and

$$G(\mathbf{r})=\left(1-\frac{1}{2}D_\perp\right)^2 e^{-1/2\mathcal{D}^{\parallel}(\mathbf{r})} \tag{13}$$

with

$$\mathcal{D}^{\parallel}(\mathbf{r})=(\tilde{S}N)^{-1}\sum_{\mathbf{k}}(1-e^{i\mathbf{k}\cdot\mathbf{r}})\frac{a_{\mathbf{k}}}{b_{\mathbf{k}}}\mathcal{L}(f_{\mathbf{k}}). \tag{14}$$

$G(\mathbf{r})$ being bounded and essentially constant for large $\mathbf{r}$, the asymptotic behavior of the correlation functions in the critical region is the same of the effective classical spin system. In particular, for $\lambda=0$, the correlation length can be simply found as $\xi(t)=\xi_{\mathrm{cl}}(t_{\mathrm{eff}})$. The result is reported in Fig. 4: as expected in a BKT transition, it displays a divergence at a temperature t_{BKT} which decreases with decreasing S, as a result of enhanced quantum fluctuations.

In Fig. 5 the in-plane normalized correlation functions $C(\mathbf{r})$ are reported for $\lambda=0.5$ and different values of the spin, as a function of the distance $\mathbf{r}$ at a fixed temperature $t=0.27$. As expected, for low spin the quantum fluctuations can be strong enough to drive the system in a disordered phase, i.e., with unbound vortices. Indeed the behavior of $C(\mathbf{r})$ shown in Fig. 5 changes drastically by decreasing the spin, decaying exponentially for $S=1/2$ and $S=1$ and as a power law for $S>5/2$.

Within the PQSCHA, the quantum renormalizations cannot modify the critical behavior of the effective classical system,[20] so that both the chirality and the BKT critical temperatures can be connected to their classical counterparts by the self-consistent relation

$$\frac{t_{\mathrm{crit}}(S,\lambda)}{t^{\mathrm{cl}}_{\mathrm{crit}}(\lambda_{\mathrm{eff}}(t_{\mathrm{crit}},S,\lambda))}=j_{\mathrm{eff}}(t_{\mathrm{crit}},S,\lambda), \tag{15}$$

which can be solved numerically. The obtained critical temperatures for $\lambda=0$ and $\lambda=0.5$ are reported for various values of the spin in Table I. In the $S=1/2$ case, although our theory begins to become unreliable when $t\lesssim0.2$, we notice that for $\lambda=0$ the extrapolated value for the chiral critical temperature, $t_c=0.193$, agrees remarkably well with those obtained by the size scaling on the quantum Monte Carlo[8] data [$t_c=0.195(1)$] and the effective field theory of Ref. 10 ($t_c\simeq0.20$).

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