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Spin-liquid phase in frustrated Heisenberg antiferromagnets

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The possible existence of nonconventional ground-state configurations for the Heisenberg antiferromagnetic model in the two-dimensional square lattice has generated great theoretical interest. Frustration caused by suitable exchange competition between spins localized on lattice sites at different distances is believed to be a possible source of the *spin-liquid* phase, where no long-range order at zero temperature is present. The early signal of this phase was provided by a divergent spin reduction, obtained by the simple spin-wave approximation, on the phase boundary between the Néel configuration and the helix configuration of the square frustrated Heisenberg antiferromagnet. Later the existence of the spin-liquid phase was investigated by renormalization-group analysis. Here we provide arguments that show that the spin-liquid phase, if it exists, cannot be located where the simple spin-wave approach suggests. This is due to crucial nonlinear contributions to the magnon self-energy. We find that the stability region of the Néel configuration is enhanced by nonlinear effects when the third-nearest-neighbor interaction is present.

The discovery of high-T_c superconductors[1] added new interest to the two-dimensional (2D) Heisenberg model because the origin of the superconductivity was related to possible nonconventional ground-state configurations of this model in the presence of antiferromagnetic coupling.[2] The square Heisenberg antiferromagnet with nearest-neighbor (NN) coupling[3] exhibits long-range order (LRO) at zero temperature for $S > \frac{1}{2}$, and well-grounded arguments[4] support the existence of LRO at $T=0$ even in the extreme quantum limit $S=\frac{1}{2}$. Frustration caused by exchange-coupling competition is the well-known origin of noncollinear configurations which are exhaustively represented by helical configurations[5] in the classical limit ($S\rightarrow\infty$). The study of the zero-temperature classical phase diagram in the parameter space of 2D models with competitive interactions up to third-nearest neighbors provided novel features as infinite degeneration lines on which the minimum-energy configurations are infinite inequivalent isoenergetic helices characterized by infinite helix wave vectors $\mathbf{Q}$. Along these degeneration lines the magnon spectrum shows soft lines.[6] The spin reduction diverges in the above 2D model as well as the thermal demagnetization does at finite temperature in the corresponding 3D tetragonal models.[6] These results, which indicate the critical dimensionality support of the LRO increase of one were obtained in classical approximation for tetragonal Heisenberg and planar models with in-plane interactions up to third-nearest neighbors, the NN interaction J_1 being ferromagnetic with a NN interplane coupling J'. The interesting phenomenology called the *degenerate helix* is also present if the NN in-plane interaction is antiferromagnetic.[7,8] Moreover, the phase boundary between the antiferromagnetic configuration and the helical configurations is characterized by a divergent spin reduction in the 2D case. This divergence was interpreted as the signal that a new phase, called *spin liquid,* intervenes between the antiferromagnetic and the helical phases.[9] We stress that such conclusions are drawn on the basis of the simple spin-wave theory so that their reliability has to be tested against quantum and thermal fluctuations, additional perturbations, doping, and so on.

Here we show that the zero-point-motion fluctuations cause drastic changes in the location of the spin-liquid phase, which is currently believed to occur around the classical helix-antiferromagnetic (H-AF) phase transition. This expectation is based on the divergence of the spin reduction caused by a k^2 behavior of the magnon dispersion curve in the long wavelength limit, so that LRO is believed to be destroyed even at zero temperature in a finite stripe encompassing the H-AF line. On the contrary we prove that this is an artifact of the linear spin-wave approximation because quantum fluctuations provide a nonzero magnon velocity restoring LRO on the classical H-AF line. Our calculation cannot be pushed onto vanishing in-plane third-nearest-neighbor interaction J_3, but for not-too-small J_3 we are able to prove that the spin-liquid phase has to be searched for well inside the region where helix configurations are stable in classical approximation.

Let us consider the magnon spectrum appropriated to a two-sublattice antiferromagnetic configuration including the first quantum correction[10]

$$(\hbar\omega_{\mathbf{k}}^{l})^2=(8J_1S)^2\left[d_{\mathbf{k}}s_{\mathbf{k}}\left(1+\frac{1}{S}\right)+\frac{1}{2NS}\sum_{\mathbf{q}}s_{\mathbf{k}}[(d_{\mathbf{k}-\mathbf{q}}-d_{\mathbf{k}})(s_{\mathbf{q}}/d_{\mathbf{q}})^{1/2}-(d_{\mathbf{q}}s_{\mathbf{q}})^{1/2}]\right.$$
$$\left.+\frac{1}{2NS}\sum_{\mathbf{q}}d_{\mathbf{k}}[(d_{\mathbf{k}-\mathbf{q}}-s_{\mathbf{k}})(d_{\mathbf{q}}/s_{\mathbf{q}})^{1/2}-(d_{\mathbf{q}}s_{\mathbf{q}})^{1/2}\right]. \quad (1)$$

For the square frustrated Heisenberg antiferromagnetic (FHA) model we have

$$d_{\mathbf{q}}=1-\tfrac{1}{2}(\cos q_x+\cos q_y)-j_2(1-\cos q_x\cos q_y)-j_3[1-\tfrac{1}{2}(\cos 2q_x+\cos 2q_y)]\,, \tag{2}$$

$$s_{\mathbf{q}}=1+\tfrac{1}{2}(\cos q_x+\cos q_y)-j_2(1-\cos q_x\cos q_y)-j_3[1-\tfrac{1}{2}(\cos 2q_x+\cos 2q_y)]\,, \tag{3}$$

where $j_2=J_2/J_1$, $j_3=J_3/J_1$. Equation (1) becomes

$$(\hbar\omega_{\mathbf{k}}^l)^2=(8J_1S)^2\left[d_{\mathbf{k}}s_{\mathbf{k}}+\frac{1}{2S}(s_{\mathbf{k}}d_{\mathbf{k}}^{\prime}+d_{\mathbf{k}}s_{\mathbf{k}}^{\prime})\right] \tag{4}$$

where

$$d_{\mathbf{k}}^{\prime}=(1-A_1)[1-\tfrac{1}{2}(\cos k_x+\cos k_y)]-j_2(1-A_2)(1-\cos k_x\cos k_y)-j_3(1-A_3)[1-\tfrac{1}{2}(\cos 2k_x+\cos 2k_y)]\,, \tag{5}$$

$$s_{\mathbf{k}}^{\prime}=(1-A_1)[1+\tfrac{1}{2}(\cos k_x+\cos k_y)]-j_2(1-A_2)(1-\cos k_x\cos k_y)-j_3(1-A_3)[1-\tfrac{1}{2}(\cos 2k_x+\cos 2k_y)] \tag{6}$$

with

$$A_1=\frac{1}{N}\sum_{\mathbf{q}}[1-\tfrac{1}{2}(\cos q_x+\cos q_y)](s_{\mathbf{q}}/d_{\mathbf{q}})^{1/2}\,, \tag{7}$$

$$A_2=\frac{1}{N}\sum_{\mathbf{q}}(1-\cos q_x\cos q_y)(s_{\mathbf{q}}/d_{\mathbf{q}})^{1/2}\,, \tag{8}$$

$$A_3=\frac{1}{N}\sum_{\mathbf{q}}[1-\tfrac{1}{2}(\cos 2q_x+\cos 2q_y)](s_{\mathbf{q}}/d_{\mathbf{q}})^{1/2}\,. \tag{9}$$

In deriving Eqs. (5) and (6) use has been made of the sum rule

$$A_1-j_2A_2-j_3A_3=\frac{1}{N}\sum_{\mathbf{q}}(d_{\mathbf{q}}s_{\mathbf{q}})^{1/2}\,. \tag{10}$$

We focus on the long-wavelength behavior of the magnon spectrum on the H-AF line $j_3=(1-2j_2)/4$:

$$\begin{aligned}(\hbar\omega_{\mathbf{k}}^l)^2=(8J_1S)^2\Bigg\{&\tfrac{1}{8}(1-2j_2)(k_x^4+k_y^4)+\tfrac{1}{2}j_2k_x^2k_y^2+\frac{1}{4S}[A_3-A_1-2j_2(A_3-A_2)](k_x^2+k_y^2)-\frac{1}{48S}\\&\times\{1-A_1-2j_2(1-A_2)-4(1-2j_2)(1-A_3)+3[A_3-A_1-2j_2(A_3-A_2)]-3(1-2j_2)(1-A_1)\}\\&\times(k_x^4+k_y^4)+\frac{1}{8S}[2j_2(2-A_1-A_2)-(A_3-A_1)+2j_2(A_3-A_2)]k_x^2k_y^2+\cdots\Bigg\}\,.\end{aligned} \tag{11}$$

We notice that the classical long-wavelength behavior is strongly modified as follows

$$\hbar\omega_{\mathbf{k}}^l=8|J_1|\sqrt{S}ck\,, \tag{12}$$

where

$$c=\tfrac{1}{2}[A_3-A_1-2j_2(A_3-A_2)]^{1/2}\,. \tag{13}$$

For instance, we find $c=0.43824$, 0.43236, 0.42767, 0.42625, 0.42601, for $j_2=0$, 0.1, 0.2, 0.25, 0.3, respectively. As anticipated, we cannot explore the vanishing j_3 limit corresponding to $j_2=0.5$ because in that limit the quantum correction diverges. For this reason we cannot perform any statement about the point $j_2=0.5$, $j_3=0$. On the contrary, for $j_3>0$ any divergence of the spin reduction is washed by the quantum fluctuations which are currently believed to play a minor role. This expectation is usually true, but it can be completely false where pathological scenarios are suggested by the classical approximation. The inclusion of the leading quantum corrections leads to the following zero-temperature expectation value of the sublattice magnetization along the quantization ζ axis

$$\langle S_f^{\zeta}\rangle=S+\frac{1}{2}-\frac{1}{2N}\sum_{\mathbf{q}}(s_{\mathbf{q}}^l/d_{\mathbf{q}}^l)^{1/2}\,, \tag{14}$$

where

$$\begin{aligned}s_{\mathbf{q}}^l=&[1+(1/2S)(1-A_1)][1+\tfrac{1}{2}(\cos q_x+\cos q_y)]-j_2[1+(1/2S)(1-A_2)](1-\cos q_x\cos q_y)\\&-j_3[1+(1/2S)(1-A_3)][1-\tfrac{1}{2}(\cos 2q_x+\cos 2q_y)]\,,\end{aligned} \tag{15}$$

$$\begin{aligned}d_{\mathbf{q}}^l=&[1+(1/2S)(1-A_1)][1-\tfrac{1}{2}(\cos+\cos q_y)]-j_2[1+(1/2S)(1-A_2)](1-\cos q_x\cos q_y)\\&-j_3[1+(1/2S)(1-A_3)][1-\tfrac{1}{2}(\cos 2q_x+\cos 2q_y)]\,.\end{aligned} \tag{16}$$

Table I gives the zero-temperature sublattice magnetization for selected values of S and j_2 on the classical H-AF line.

TABLE I. Sublattice magnetization at zero temperature [see Eq. (14)] for selected values of $j_2=J_2/J_1$ and the spin S along the classical H-AF transition line $j_3=(1-2j_2)/4$.

j_2	$S=\frac{1}{2}$	$S=1$	$S=\frac{3}{2}$	$S=2$
0	0.260	0.649	1.074	1.516
0.1	0.255	0.639	1.059	1.498
0.2	0.253	0.630	1.045	1.479
0.25	0.254	0.626	1.038	1.470
0.3	0.256	0.624	1.033	1.462

We conclude that previous evaluations concerning the location of the spin-liquid phase are unreliable at least for $j_3 \geq 0.1$. It is clear that the linear spin-wave theory is unreliable where crucial nonlinear (quantum) effects bring dramatic corrections to the spin-wave energy. Our result is in contrast not only with the simple spin-wave argument,[9] but even with the renormalization-group analysis[11] recently performed for the square FHA model. Indeed, as one can see in Fig. 2 of Ref. 11 the spin-liquid phase region is qualitatively the same as that obtained by linear spin-wave approach,[9] except that it is sensibly increased inside the classical Néel region. We think that this is a spurious result which comes from the structure of the effective action assumed in Eq. (3.10) of Ref. 11. We notice that the *spatial* part of that action vanishes on the classical H-AF phase boundary, so that higher-order contributions are crucial in order to treat conveniently the vicinity of the H-AF line. This is analogous to the simple spin-wave approach that neglects nonlinear quantum contributions which enter dramatic corrections to the spin-wave velocity where the linear contributions vanish. In conclusion, we do not exclude that the spin-liquid phase exists, but we believe that its existence region has to be searched for inside the region where the helix configuration is found in classical approximation, at least for not-too-small j_3 coupling. On the other hand, we stress that a first-order H-AF phase transition could prevent the onset of the spin-liquid phase. Notice that this possible scenario, which has not been investigated for the square FHA model, has been found in the same model with a ferromagnetic NN coupling.[12,13] Just close to the triple point where ferromagnetic, antiferromagnetic, and helical phases coexist, the F-H phase transition has been proven to be changed from continuous to first order owing to long-wavelength quantum fluctuations. Anyway, we estimate the possible location of the AF spin-liquid phase boundary for the FHA model, assuming that this model does not undergo a first-order phase transition. This assumption seems reliable because our calculation involves finite values of the third-nearest-neighbor coupling j_3 where the influence of the quantum fluctuations is expected to be less dramatic as one moves away from the $j_2=0.5$, $j_3=0$ triple point. In Fig. 1 we show for selected values of S the line in the parameter space where the exchange competition cancels the quantum contribution to

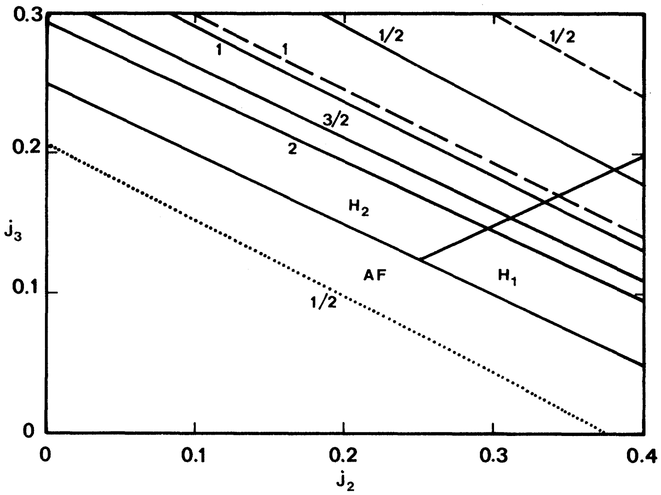

FIG. 1. AF, H_1, H_2 indicate the classical existence regions of the Néel and two helix phases. Equations of AF-H and H_1-H_2 transition lines are $j_3=(1-2j_2)/4$ and $j_2=2j_3$, respectively. Solid and dashed lines are the AF spin-liquid transition lines and the locus where the AF magnon velocity vanishes, respectively. The labels refer to $S=\frac{1}{2}$, 1, $\frac{3}{2}$, 2, respectively. For $S=\frac{3}{2}$ and 2 solid and dashed curves are not distinguishable. The dotted line is the AF spin-liquid transition line obtained by the simple spin-wave approximation for $S=\frac{1}{2}$.

the magnon velocity given by Eq. (5), so leading to a soft behavior of the long-wavelength magnon dispersion curve that causes a catastrophic spin reduction. In Fig. 1 the line where the *spin-liquid* phase onsets is shown. As one can see, this line occurs before the line where the spin-wave velocity vanishes is reached. From this point of view the behavior is similar to the one suggested by the simple spin-wave approach, but a striking difference appears because nonlinear quantum contributions push such a line inside the region of existence of the helix phase obtained in classical approximation ($S \rightarrow \infty$) in contrast with previous results[9,11] that provide the existence of the spin-liquid phase inside the AF classical region.

Note added. We have recently received a copy of unpublished work by Chubukov[14] concerning the same problem we have studied in our paper. The emphasis of the work of Chubukov is devoted to the vanishing J_3 region where a first-order transition between the Néel and the columnar phases is announced. Our approach is valid for finite J_3 and we cannot achieve the $J_3=0$ limit. Even if our results agree qualitatively with the Chubukov expectations for finite J_3, it is not clear to us at this time how the author obtains Eq. (4′) characterizing the instability point of the Néel spin wave. Indeed, our controlled approach provides divergent results for $J_2=J_1/2$, $J_3=0$. Notice that this drawback can be healed only by some *ad hoc* heuristic ansatz. We think that the neighborhood of such a point deserves further theoretical effort.

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