RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 38, NUMBER 13 1 NOVEMBER 1988

Possible spin-liquid state at large S for the frustrated square Heisenberg lattice

P. Chandra
Joseph Henry Laboratories of Physics, Jadwin Hall, Princeton University, Princeton, New Jersey 08544
and Department of Physics, University of California at Santa Barbara, Santa Barbara, California 93106

B. Doucot
Joseph Henry Laboratories of Physics, Jadwin Hall, Princeton University, Princeton, New Jersey 08544
(Received 9 May 1988; revised manuscript received 30 June 1988)

We present conventional spin-wave calculations for a frustrated antiferromagnetic Hamiltonian on a square lattice. We find that quantum fluctuations can destabilize the classical ordered ground state, even at large S, for large enough values of frustration. This instability is likely to generate a spin liquid, providing the first example of a resonating-valence-bond ground state in two dimensions.

The recent discovery of the layered oxide high-temperature superconductors has stimulated considerable interest in two-dimensional antiferromagnetic systems.[1] Anderson[2] has proposed that for small spin values, strong quantum fluctuations may generate a novel spin-liquid ground state with no long-range order. The relevant geometry for the Cu-O planes is the square lattice; many studies indicate that there exists a finite zero-temperature staggered magnetization.[3–6] However, it has also been shown[7] that the energy difference between ordered and disordered states in this configuration is very small (~0.2%); therefore, it is of great interest to test the stability of the long-range Néel order.[8] In this Rapid Communication we introduce frustration as a perturbation and use conventional spin-wave theory[3] to study the first quantum corrections to the ground-state staggered magnetization. We find that there exists a small but finite region in parameter space (spin and frustration) where zero-point fluctuations are strong enough to melt any ordered state. Though this approach cannot lead to a rigorous existence proof for a spin liquid, in analogy with the Ginzburg criterion, it does indicate that the assumption of an ordered ground state is no longer consistent for large values of the frustration parameter α.

We consider the square-diagonal Heisenberg lattice

$$H=J_1\sum_{\langle i,j\rangle} S_iS_j+J_2\sum_{\langle i,i'\rangle} S_iS_{i'}+J_2\sum_{\langle j,j'\rangle} S_jS_{j'}\,, \qquad (1)$$

shown in Fig. 1 where J_1 and J_2 correspond to the nearest- and next-nearest-neighbor bonds, respectively. Here the subscript $i(j)$ denotes sites on the sublattice $A(B)$. The advantage of such a model is that it retains the square lattice symmetry while simultaneously exhibiting frustration. If we take $\alpha\equiv J_2/J_1$ then classically ($S\to\infty$) for $\alpha<\frac{1}{2}$ the ground state has the conventional Néel order with two sublattices. When $\alpha>\frac{1}{2}$, the same classical limit yields a ground state where the previous sublattices are decoupled and each has antiferromagnetic order as shown in Fig. 1. At $\alpha=\frac{1}{2}$ any state with total spin equal zero for an elementary square is a ground state; this includes the two configurations discussed above as well as many others with no long-range order.

We can study zero-point fluctuations about the two possible ordered states in the large S limit using conventional spin-wave theory.[3] $1/S$ corrections to states 1 and 2 yield the following sublattice magnetization per spin, respectively:

$$(S_z)^1=(S+\tfrac{1}{2})-\frac{1}{8\pi^2}\int d^2k\frac{1-\alpha(1-\eta_k)^2}{\{[1-\alpha(1-\eta_k)]^2-\gamma_k^2\}^{1/2}}\,, \qquad (2)$$

and

$$(S_z)^2=(S+\tfrac{1}{2})-\frac{1}{16\pi^2}\int d^2k\frac{1+\lambda\gamma_{k,y}}{[(1+\lambda\gamma_{k,y})^2-(\eta_k+\lambda\gamma_{k,x})]^{1/2}}+\frac{1-\lambda\gamma_{k,y}}{[(1-\lambda\gamma_{k,y})^2-(\eta_k-\lambda\gamma_{k,x})]^{1/2}}\,, \qquad (3)$$

where $\gamma_{kx}=\cos k_xa$, $\gamma_{ky}=\cos k_ya$, $\gamma_k=\frac{1}{2}(\cos k_xa+\cos k_ya)$, $\eta_k=\cos k_xa\cos k_ya$, and $\lambda=1/2\alpha$. We note that for $\alpha=0$, (2) and (3) lead to the Anderson result[3] for the square lattice, as expected. The $O(1/S)$ spin-wave theory predicts a vanishing order parameter along two lines in the $1/S$ vs α phase diagram as shown in Fig. 2. Based on this result, it is reasonable to speculate that the ground state in the intermediary region will be nonmagnetic and will therefore be a spin liquid even at large values of S. Corrections to $O(1/S^3)$ for $\alpha<\frac{1}{2}$ indicate that this liquid state is preserved,[9] and therefore we have reason to believe that this will be the case for all orders in

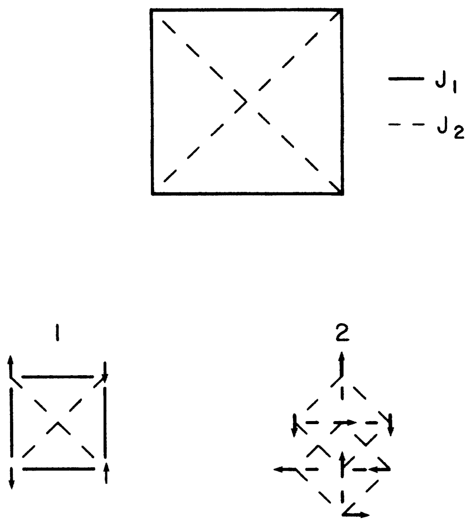

FIG. 1. The frustrated square lattice with couplings J_1 and J_2 shown with classical ground states 1 and 2 as defined in text.

$1/S$. Furthermore, the quantum fluctuations diverge for $\alpha \rightarrow \frac{1}{2}$; the asymptotic behavior for states 1 and 2 is

$$\frac{1}{S} \sim \frac{1}{\ln|\frac{1}{2} - \alpha|}, \tag{4}$$

and

$$\frac{1}{S} \sim (\alpha - \tfrac{1}{2})^{1/2}, \tag{5}$$

respectively. For values far from $\alpha = \frac{1}{2}$, the liquid region is wider for $\alpha < \frac{1}{2}$ than for $\alpha > \frac{1}{2}$; this asymmetry corresponds to the fact that only quantum fluctuations melt state 2, whereas state 1 is already destabilized by frustration at the classical level. We emphasize that both the continuous symmetry and the low dimensionality of this system are crucial to the presence of the liquid state at large S; soft modes contribute to the large amplitude of the zero-point motion at long wavelengths and yield an infrared divergence for dimensions $d \leq 2$.

We would like to stress the important fact that classically ($S = \infty$) the system at $\alpha = \frac{1}{2}$ is *not* strictly disordered; the constraint that each square has total spin zero induces a sort of rigidity. In particular, a generic ground state may be written as

$$\sigma(x,y) = \epsilon(y)(-1)^x, \quad \epsilon(y) = \pm 1 \tag{6}$$

(or $x \leftrightarrow y$), where $\sigma(x,y) = \pm 1$ is the projection of the classical spin along the z direction, and $\epsilon(y)$ is an arbitrary function. This shows that there is no residual finite entropy at $T = 0$ for $\alpha = \frac{1}{2}$. However, our claim here is that as $\alpha \rightarrow \frac{1}{2}$, leading quantum fluctuations become very soft, driving the system away from the classical fixed point. We emphasize that the conjectured spin-liquid phase is then quite different from the classical ($S = \infty$) system at $\alpha = \frac{1}{2}$. From our previous calculations, we expect that for any of the possible classical ground states, the relevant order parameter is also destroyed by the leading infrared fluctuations.

In order to give further support to the existence of a spin-liquid ground state, we need to show explicitly that an order state with uniform twist (Fig. 3) cannot be stabilized by quantum fluctuations in the vicinity of $\alpha = \frac{1}{2}$. Such a modulated phase exists, for example, near the frustrated point in both the asymmetric $p \geq 3$ clock[10] and anisotropic nearest-neighbor Ising (ANNI)[11] models, and one might speculate that thermal fluctuations in these classical discrete Hamiltonians could have some analogy with the zero-point motion in our quantum Heisenberg system.

We have calculated $1/S$ corrections to the ground-state energy for an ordered state of arbitrary twist. We recall that for $\alpha < \frac{1}{2}$ and $\alpha > \frac{1}{2}$ there exists only one twist vector Q in the ground state of the classical system ($Q = 0$ and $Q = \pi$, respectively). Spin-wave theory indicates that twisted states with Q other than the relevant classical value are all unstable; fluctuations about these states lead to the presence of modes with negative energy. At $\alpha = \frac{1}{2}$ all twisted states are allowed, though there exists only one global energy minimum at $Q = 0$. This reflects the classi-

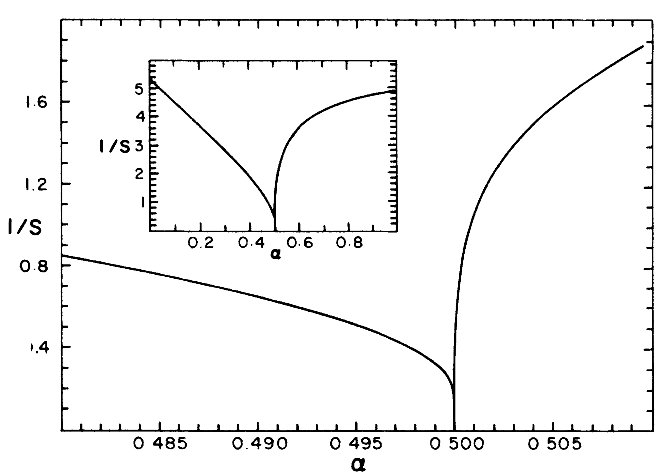

FIG. 2. Phase diagram obtained by comparison of the classical sublattice magnetization and the first quantum corrections.

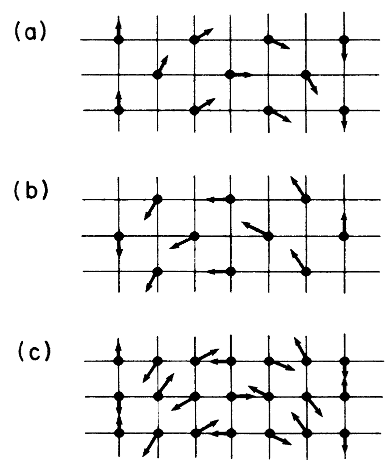

FIG. 3. An example of an ordered state with a twist along the x axis; here $Q = \pi/6$. (a) A sublattice; (b) B sublattice; (c) A and B sublattices.

cal degeneracy at $\alpha=\frac{1}{2}$. In order to check our result that the quantum fluctuations do not select Q states different from those expected classically, we have investigated an extension of this model where a twisted ground state may occur at the classical level. Specifically, we introduce next-nearest-neighbor couplings βJ_1 along the x axis. The classical phase diagram at $T=0$ for this model is shown in Fig. 4. We note the presence of a twisted ground state for

$$\beta \geq \frac{|2\alpha-1|}{4} \left[\cos Q = \frac{1-2\alpha}{4\beta} \right] .$$

Spin-wave theory can also be applied in this $\alpha\beta$ model, and yields the following frequencies:

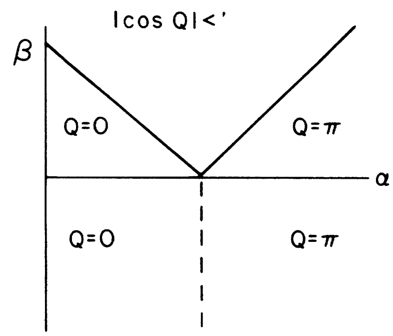

FIG. 4. Classical phase diagram for the $\alpha\beta$ model.

$$(w_k^A)^2 = [1+(1-2\alpha)\cos(Q) - \beta\cos(2Q) + \gamma_{k,x} + \gamma_{k,y} + 2\alpha\eta_k + \beta\gamma_{2k,x}]$$
$$\times [1+(1-2\alpha)\cos(Q) - \beta\cos(2Q) - \gamma_{k,x}\cos(Q) - \gamma_{k,y} + 2\alpha\eta_k\cos(Q) + \beta\cos(2Q\gamma_{2k,\gamma})] , \quad (7a)$$

$$(w_k^B)^2 = (w_{k+(\pi,\pi)}^A)^2 , \quad (7b)$$

where the notation is the same as before. Again, for $\alpha \neq \frac{1}{2}$, the only stable states correspond to the classical values of Q. At $\alpha=\frac{1}{2}$, the two states $Q=\pi/2$ and $Q=3\pi/2$ are stable and degenerate for $\beta>0$; the degeneracy reflects a discrete symmetry of the twisted state $Q \rightarrow -Q$. Though both thermal and quantum fluctuations lift degeneracies which may occur in their absence,[12] these results indicate an important difference between these phenomena. Because of entropy considerations, finite-temperature effects may modify the symmetry of the $T=0$ ground state and may even create a new length scale in the system.[11] It appears, however, that quantum fluctuations will not select a state that is classically not permitted since they act to restore the rotational invariance of the system.

In conclusion, it seems very likely that a spin liquid exists at zero temperature in a finite region of the phase diagram (even at large S) for the frustrated square Heisenberg model. If the ground state does not break translational invariance (dimerized case), this would provide the first example of a resonating-valence-bond (RVB) ground state for a two-dimensional isotropic Hamiltonian. At this stage, it is not possible to distinguish between the "long bond" RVB discussed by Anderson,[2] and the "short bond" RVB of Kivelson, Rokhsar, and Sethna.[13] Numerical studies for the spin-$\frac{1}{2}$ case are now in progress to investigate this new phase.[14,15] Naturally, there remain many open questions including, for example, the order of the transition, the nature of elementary excitations in the liquid state, and the behavior of the spin-spin correlation functions. It would also be interesting to investigate the continuum limit in this model and to determine whether or not it can be described by the disordered fixed point in the O(3) σ model discussed by Chakravarty, Halperin, and Nelson.[6]

Note added. After completion of this work, we received a preprint by Ioffe and Larkin[16] where they derived an effective action to describe the low-energy behavior of the same frustrated model as studied here. Their conclusions are consistent with ours; in particular, they find a spin-liquid phase, even at large S, for large values of the frustration parameter α.

The authors would like to thank B. S. Shastry for suggesting this project, and D. A. Huse for many valuable discussions concerning the nature of the twisted state. We have also benefited from conversations with S. Chakravarty, P. Coleman, S. Liang, and S. Sondhi. One of us (P.C.) is grateful to P. W. Anderson and Princeton University for their hospitality when this work was performed, and acknowledges receipt of financial support from the University of California. This work was supported by U.S. Department of Energy Grant No. DE-FG03-84ER45108 (University of California at Santa Barbara), National Science Foundation Grants No. DMR-8518163 (Princeton University) and No. PHY82-17853, supplemented by funds from the National Aeronautics and Space Administration (University of California at Santa Barbara).

[1]For example, *Novel Superconductivity, Proceedings of the International Workshop on Novel Mechanisms of Superconductivity, Berkeley, California, 1987,* edited by S. A. Wolf and V. Z. Kresin (Plenum, New York, 1987).

[2]P. W. Anderson, Science **235**, 1196 (1987); in *Frontiers and Borderlines in Many Particle Physics, Proceedings of the Enrico Fermi International Summer School of Physics,* edited by J. R. Schrieffer and R. A. Broglia (North-Holland, Amsterdam, in press).

[3]P. W. Anderson, Phys. Rev. **86**, 694 (1952); R. Kubo, *ibid.* **87**, 568 (1952); T. Oguchi, *ibid.* **117**, 117 (1960).

[4]J. D. Reger and A. P. Young, Phys. Rev. B **37**, 5493 (1988); **37**, 5978 (1988).

[5]D. A. Huse and V. Elser (unpublished).

[6]S. Chakravarty, B. I. Halperin, and D. R. Nelson, Phys. Rev. Lett. **60**, 1057 (1988).
[7]S. Liang, B. Doucot, and P. W. Anderson (unpublished).
[8]H. Yokoyama and H. Shiba, J. Phys. Soc. Jpn. **56**, 1490 (1987); C. Gros, R. Joynt, and T. M. Rice, Phys. Rev. B **36**, 381 (1987).
[9]S. Chakravarty (private communication).
[10]S. Ostlund, Phys. Rev. B **24**, 398 (1981).
[11]J. Villain and P. Bak, J. Phys. (Paris) **42**, 657 (1981), and references therein.
[12]J. Villain, R. Bideaux, J. P. Carton, and R. Comte, J. Phys. (Paris) **41**, 1263 (1980); C. L. Henley, J. Appl. Phys. **61**, 3962 (1987).
[13]S. Kivelson, D. Rokhsar, and J. P. Sethna, Phys. Rev. B **35**, 8865 (1987).
[14]S. Liang (private communication).
[15]S. Sondhi (private communication).
[16]L. B. Ioffe and A. I. Larkin (unpublished).