Random frustration in a two-dimensional spin-$\frac{1}{2}$ Heisenberg antiferromagnet

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The square-lattice spin-1/2 antiferromagnet containing a dilute concentration δ of randomly placed ferromagnetic nearest-neighbor bonds is studied at low-temperature via nonlinear σ-model techniques and by exact diagonalization. We generally find that long-range Néel order is destroyed above a critical strength in the defective ferromagnetic exchange-coupling constant given by $|K_c|/J \sim \delta^{-1/2}$. We also observe large statistical fluctuations both in the spin stiffness and in the antiferromagnetic structure factor near this critical point, suggesting the onset of a spin-glass phase.

It is now established that the spin-$\frac{1}{2}$ Heisenberg model on the square lattice with nearest-neighbor antiferromagnetic coupling displays long-range Néel order at zero temperature, along with a correlation length that diverges exponentially at low temperature.[1] This model, in fact, successfully accounts for the long-range two-dimensional (2D) antiferromagnetic correlations that are observed in "parent" compounds to high-temperature superconductors such as La_2CuO_4 at temperatures just above the transition temperature to three-dimensional Néel ordering.[1] However, the latter antiferromagnetic insulating state disappears rapidly upon hole doping, where it is followed eventually by the appearance of the superconducting phase.[2]

Aharony *et al.* have proposed that such doping results in a frustrating effective ferromagnetic coupling between the nearest-neighbor copper spins situated near the hole, which is presumed to lie statically at the intermediate oxygen superexchange site.[3] (A three-band Hubbard model is implicit in this approach.) The observation of a disordered spin state via neutron-scattering experiments in the $La_{2-x}Sr_xCuO_4$ series for temperatures below 100 K, and in the doping range $0.015 < x < 0.05$, supports this scenario.[2] In this paper, we elucidate the theoretical consequences of the former proposal by studying the low-temperature properties of a spin-$\frac{1}{2}$ nearest-neighbor Heisenberg antiferromagnet on the square lattice with a quenched random distribution of dilute ferromagnetic nearest-neighbor bonds. In particular, we first consider the corresponding classical problem at nonzero temperature via the nonlinear σ model,[4] which is known to correctly describe the pure 2D antiferromagnet.[1] We then compute the spin stiffness, as well as various spin correlations, of the full quantum problem at zero temperature by exact diagonalization on a 4×4 lattice using the Lanczos technique.[5] For a fixed concentration δ of frustrating ferromagnetic bonds, both studies find that long-range Néel order disappears on average at a critical strength in the nearest-neighbor ferromagnetic exchange-coupling constant given by $|K_c|/J \sim \delta^{-1/2}$. In the case of antiferromagnetic defective bonds, however, the exact diagonalization study finds no evidence for the destruction of long-range Néel order, as one intuitively expects. In addition, the statistical fluctuations shown by both the spin stiffness and the antiferromagnetic structure factor obtained in the exact diagonalization study become extremely large near the former critical point, suggesting the onset of a spin-glass phase.[3] Last, our numerical calculations also reveal that uniform chiral spin fluctuations[6] are enhanced by the introduction of either antiferromagnetic or ferromagnetic defective bonds. A discussion of these results within the context of the low-temperature magnetic phase diagram of $La_{2-x}CuO_4$ is given at the end of the paper.

Consider a nearest-neighbor spin-$\frac{1}{2}$ Heisenberg model Hamiltonian,

$$H=\sum_{\langle ij\rangle} J_{ij}\mathbf{S}_i\cdot\mathbf{S}_j\ , \tag{1}$$

on the square lattice, with a dilute concentration δ of randomly distributed bonds, $J_{ij}=K<0$, lying within a uniform antiferromagnetic background with $J_{ij}=J>0$.[7] Clearly then, the average value and the standard deviation of the exchange-coupling constant per *bond* are given by

$$J_0=K\delta+J(1-\delta) \tag{2a}$$

and

$$J_1=|J-K|[(1-\delta)\delta]^{1/2}\ , \tag{2b}$$

respectively. Below, we analyze the classical nonlinear σ model for the present antiferromagnet with random frustration and find that the destruction of long-range Néel

order at low temperature on average is given by the Lindemann-like criterion $J_0^2 \sim J_1^2$.

Classical nonlinear σ model. Suppose that long-range Néel order exists in the ground state of Hamiltonian (1). In analogy with the treatment of the pure 2D quantum antiferromagnet,[1] we consider then the classical 2D nonlinear σ model[4] with a randomly fluctuating local spin-stiffness field, which presumably mimics the random frustration present in Hamiltonian (1). The partition function for this model may be expressed as

$$Z=\int \mathcal{D}\mathbf{n}(\mathbf{x})\mathcal{D}\lambda(\mathbf{x}')\exp(-\beta E)\ , \tag{3}$$

with an energy functional given by

$$E=\frac{s^2}{2}\int d^2x\{[J_0+J_1(\mathbf{x})]|\nabla\mathbf{n}(\mathbf{x})|^2 + i\lambda(\mathbf{x})[|\mathbf{n}(\mathbf{x})|^2-1]\}\ , \tag{4}$$

such that $J_1(\mathbf{x})$ is a randomly fluctuating local spin-stiffness field with zero average value. Here, $\mathbf{n}(\mathbf{x})$ is the Néel-order parameter constrained to unit modulus by integration over the Lagrange-multiplier field $\lambda(\mathbf{x})$, while $s=\frac{1}{2}$ is the spin per site. (Quantum effects renormalize down s^2 in the pure case.[1,5,8]) Fourier transformation of all the fields above yields a fluctuation contribution to the energy (4) equal to

$$E_1=-\frac{s^2}{2}\sum_{\mathbf{q}}\sum_{\mathbf{k}}\sum_{\mathbf{k}'}J_1(\mathbf{q})[\mathbf{n}(\mathbf{k})\cdot\mathbf{n}(\mathbf{k}')] \times(\mathbf{k}\cdot\mathbf{k}')\delta(\mathbf{k}+\mathbf{k}'+\mathbf{q})\ . \tag{5}$$

For the sake of calculational convenience, let us now generalize the three-dimensional Néel-order parameter to an N-dimensional vector field. In the large-N limit we may ignore fluctuations in the Lagrange-multiplier field and take $\lambda(\mathbf{x})=\lambda_0$.[4,8] If the effect of fluctuation energy (5) is accounted for within the coherent-potential approximation (CPA),[9] which ignores backscattering, then the Greens function for the Néel-order parameter field is given by $\langle n_\alpha(\mathbf{k})n_\beta(-\mathbf{k})\rangle=G(\mathbf{k})\delta_{\alpha\beta}$, with

$$G(\mathbf{k})=(\beta s^2)^{-1}[J_0k^2+i\lambda_0-\Sigma(\mathbf{k})]^{-1}\ , \tag{6}$$

where the self-energy correction is self-consistently determined by

$$\Sigma(\mathbf{k})=\frac{1}{2}\beta s^2a^2J_1^2\int\frac{d^2k'}{(2\pi)^2}(\mathbf{k}\cdot\mathbf{k}')^2G(\mathbf{k}')\ . \tag{7}$$

Above, it has been implicitly assumed that $\langle J_1(\mathbf{x})J_1(\mathbf{x}')\rangle=2a^2J_1^2\delta(\mathbf{x}-\mathbf{x}')$, where the constant a corresponds to the lattice spacing of the original Heisenberg model (1). In general, the constraint that the Néel-order parameter be of unit modulus must also be satisfied;[4] i.e., we have that $1=\langle\mathbf{n}^2\rangle=N\int(2\pi)^{-2}d^2kG(\mathbf{k})$. Substitution of the ansatz $\Sigma(\mathbf{k})=\gamma J_0k^2$ into both this constraint and Eq. (7) yields the relationships

$$(1-\gamma)X=\ln(k_D^2\xi^2+1) \tag{8a}$$

and

$$(1-\gamma)\gamma=Y[1-(k_D\xi)^{-2}\ln(k_D^2\xi^2+1)]\ , \tag{8b}$$

respectively, where the antiferromagnetic correlation length ξ is defined by $i\lambda_0=(1-\gamma)J_0\xi^{-2}$, where $X=4\pi s^2J_0/NT$, and where $Y=(k_D^2a^2/4\pi)(J_1^2/4J_0^2)$. Here, k_D represents the momentum integration cutoff that we henceforth take to satisfy $k_D^2a^2/4\pi=a^2\int_0^{k_D}(2\pi)^{-2}d^2k=1$, by correspondence with the first Brillouin zone of the square lattice.

To begin the analysis of the above mean-field equations, we consider first the low-temperature regime, such that $X\gg1$. Then Eq. (8a) implies that the correlation length diverges exponentially, like in the pure system,[1,4] if $\gamma<1$. Hence, by Eq. (8b) we have that

$$(1-\gamma)\gamma\to Y \tag{9}$$

in the case. Yet in the dilute limit $\delta\to0$, we also have that $Y\ll1$ by Eq. (2b). Hence, (9) implies that $\gamma\cong Y$. This result in conjunction with (8a) yields, therefore, that the correlation length is approximately given by $k_D^2\xi^2=e^{(1-Y)X}$ at low temperature. Within CPA, therefore, the effect of dilute random fluctuations in the local spin stiffness is to reduce the effective spin stiffness connected with long-range Néel order[1,4] by a factor of $1-Y=1-J_1^2/4J_0^2$. On the other hand, (9) also yields that the parameter Y has a maximum value of $\frac{1}{4}$ at $\gamma_c=\frac{1}{2}$. The CPA thus also results in a critical ferromagnetic coupling constant, $K_c<0$, below which solutions to mean-field Eqs. (8a) and (8b) possessing long-range Néel order no longer exist, determined by the condition $J_0^2=J_1^2$. Substitution of expressions (2a) and (2b) into this condition then gives

$$|K_c|_{\mathrm{CPA}}\cong J/\delta^{1/2} \tag{10}$$

in the dilute limit. The exact solution for the critical value of the ferromagnetic exchange-coupling constant within CPA is displayed in Fig. 1 as a function of concentration. It is important to remark that the spin stiffness

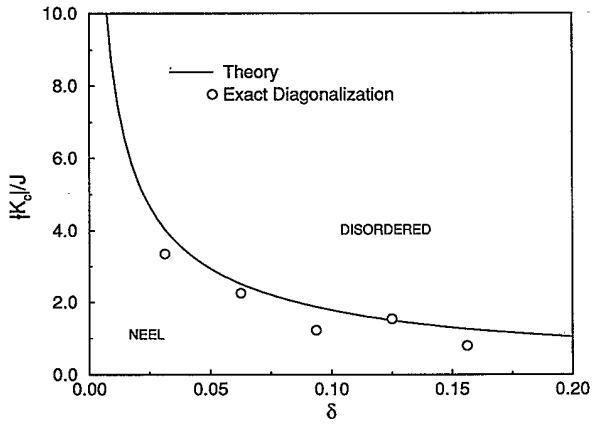

FIG. 1. The solid line given by the solution to the quadratic equation $J_0^2=J_1^2$ [see Eqs. (2a) and (2b)] delineates the point beyond which long-range Néel order is no longer possible in the present σ-model calculation. The circles correspond to the point at which the spin stiffness vanishes on average in the 4×4 exact diagonalization study for $N_b=1,2,\ldots,5$ ferromagnetic bonds, with the concentration determined by $\delta=N_b/32$.

precisely at this critical point does *not* vanish, but is reduced by a factor of $1-\gamma_c=\frac{1}{2}$ with respect to stiffness of the pure system. Second, a straightforward analysis of mean-field Eq. (8a) and (8b) demonstrate that the paramagnetic solution $\xi\to 0$ is possible either at high temperatures or when the spin stiffness vanishes. In particular, one finds that $\gamma\to\frac{1}{2}XY$ and $k_D^2\xi^2\to(1-\frac{1}{2}XY)X\ll 1$, which implies either that $X\ll 1$ or that $\gamma\to\frac{1}{2}XY\to 1$. Last, it can also be shown that no zero-temperature solution to mean-field Eqs. (8a) and (8b) exists that has a nonzero yet finite correlation length.

Exact Diagonalization. In Ref. 5, the authors have implemented a direct method for calculating the spin stiffness of generic spin-$\frac{1}{2}$ Heisenberg models (1) by exact diagonalization of small clusters with spin-dependent twisted boundary conditions in the $S_z=0$ subspace.[10] The method determines the spin stiffness of the ferromagnet to an accuracy of 1%, while it is accurate to within 10% in the case of the antiferromagnetic chain.[5] We have applied this technique to the present Hamiltonian (1) describing dilute randomly placed defective bonds within a background of homogeneous antiferromagnetic bonds on a 4×4 square-lattice. Figure 2 displays both the average and the standard deviation of the so-determined spin stiffness for 100 randomly chosen configurations of $N_b=2,4$ defective bonds as a function of the coupling constant K. The first observation to be made is that the average spin stiffness decreases rapidly to zero with increasing ferromagnetic coupling,[11] $|K|$, while it remains positive if the defective bonds are antiferromagnetic. The critical value of K at which this average value vanishes is plotted in Fig. 1 as circles for the case of $N_b=1$, 2, 3, 4, and 5 defective bonds. The agreement with the previous classical nonlinear σ-model results is quite good, indicating that quantum effects do not play an important role in the destruction of long-range Néel order by such random frustration.

The next important feature to observe is that the standard deviation of the spin stiffness obtained via the Lanczos study, in contrast to its average value, increases dramatically with increasing strength of the ferromagnetic bonds (see the inset of Fig. 2). The presence of such large statistical fluctuations in the spin stiffness supports the claim that a spin-glass phase appears once long-range Néel order is destroyed, as was suggested by Aharony *et al.*[3] Such a state is also consistent with our findings that the ground-state obtained in the present exact-diagonalization study is a spin singlet in the transition regime, while the first excited state in this regime has spin-1. Last, we mention that recent theoretical studies of quantum Heisenberg models (1) with random *infinite-range* defective bonds also find evidence for spin-glass behavior.[12]

To complete our numerical investigation, we have measured the spin-spin correlation function $\langle \mathbf{S}_i\cdot\mathbf{S}_j\rangle$ for the ground-state of Hamiltonian (1) obtained by the Lanczos technique. Figure 3 displays both the average and the standard deviation of its Fourier transform, $S(\mathbf{q})$, at the antiferromagnetic point $\mathbf{q}=(\pi,\pi)$ for 100 randomly chosen configurations of systems with $N_b=2,4$ defective bonds as a function of the coupling constant K. Contrary to the spin stiffness (Fig. 2), we see that both ferromagnetic and antiferromagnetic defective bonds suppress long-range antiferromagnetic spin correlations. The fact that $S(\pi,\pi)$ remains positive near the critical point where the spin stiffness vanishes on average could be a finite-size effect.[13] Figure 3 also shows that the standard deviation of $S(\pi,\pi)$, which is a type of Binder spin-glass order parameter,[14] increases dramatically upon the introduction of frustration, eventually saturating to a value on the order of the structure factor itself. If we presume that the spin-glass onset point coincides with the half-maximum of this standard deviation, then we obtain critical values for the defective exchange-coupling constant of $|K_c|\lesssim 2,4$ in the cases of four and two frustrating bonds, respectively. This is consistent with the point at which long-range Néel order is destroyed, as indicated in Fig. 1, and with the proposal that spin-glass order follows.[3] We

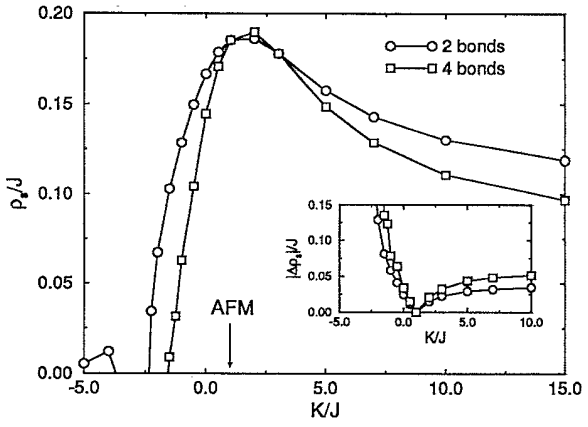

FIG. 2. Shown is the average spin stiffness for $N_b=2,4$ defective bonds as a function of the defect exchange-coupling constant K. The standard deviation of the spin stiffness is displayed in the inset.

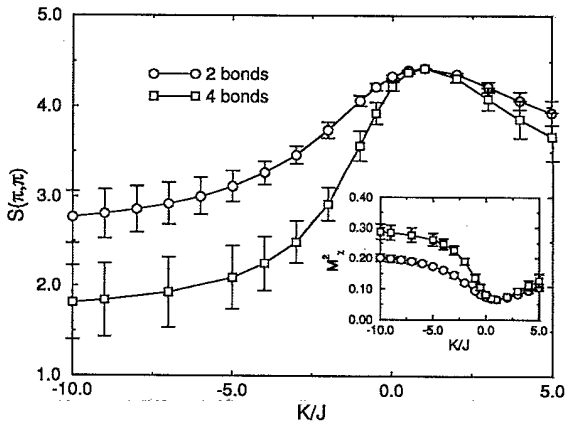

FIG. 3. The average long-range antiferromagnetic spin correlations are plotted as a function of the defect-coupling constant K for $N_b=2,4$ defective bonds, while the corresponding plot for the squared uniform chiral moment is shown in the inset. Error bars represent the standard deviation in each quantity.

have also computed the average of the squared uniform chiral moment, which we define on an arbitrary plaquette of points $i=1,2,3,4$ by

$$M_\chi^2=[\mathbf{S}_1\cdot(\mathbf{S}_2-\mathbf{S}_4)\times\mathbf{S}_3+\mathbf{S}_2\cdot(\mathbf{S}_3-\mathbf{S}_1)\times\mathbf{S}_4]^2 .$$

A straightforward yet tedious calculation reveals the remarkable identity that $M_\chi^2=\frac{1}{2}(1-P_{13}P_{24})$, where P_{ij} denotes the spin-exchange operator between sites i and j. (Notice that the Néel configuration is a null eigenstate of this operator.) Our results for this chiral moment are plotted in the inset to Fig. 3. In contrast to the behavior shown by the antiferromagnetic correlations $S(\pi,\pi)$, we observe that M_χ^2 generally increases with increasing strength in both antiferromagnetic and ferromagnetic defective bonds. It is worth mentioning here that staggered chiral spin fluctuations are exponentially suppressed at low temperature in the pure quantum-renormalized 2D Néel phase.[8]

In conclusion, both the nonlinear σ model and direct calculations of the spin stiffness for 2D spin-$\frac{1}{2}$ Heisenberg models based on exact diagonalization of a 4×4 lattice with twisted boundary conditions[5,10] show evidence for the rapid destruction of long-range Néel order at low temperature induced by the presence of dilute ferromagnetic bonds. In addition, the appearance of large statistical fluctuations in the latter spin stiffness near the point where its average value vanishes support the claim that the disordered phase is a spin glass.[3] Hence, the low-temperature phase diagram of our Hamiltonian (1) appears to be quite different from that of the *pure* quantum nonlinear σ model in $2+1$ dimensions,[1] which has recently been discussed in connection with the high-temperature magnetic behavior displayed by the doped "parent" compounds to high-T_c superconductors.[15] Last, the low-temperature Néel phase in $La_{2-x}CuO_4$ disappears at a hole concentration of $x\cong0.015$.[2] If we suppose that each of these holes corresponds to a ferromagnetic bond in the present model (1),[3] then a critical concentration $\delta=x/2=0.0075$ of such bonds implies a ferromagnetic exchange-coupling constant of $|K|\sim10J$ by the previous CPA result (10). The latter value is consistent with simple microscopic estimates based on direct copper-oxygen exchange in the oxide superconductors.[3] Furthermore, inversion of Eq. (10) shows that the critical concentration for the destruction of long-range Néel order is very sensitive to $|K|/J$, suggesting that the low-temperature magnetic phase diagram of the oxide superconductors could be sensitive to pressure. Last, we emphasize that these results indicate that long-range Néel order exists up to a *nonzero* critical concentration of frustrating bonds, contrary to statements in the literature suggesting that any amount of random frustration is sufficient to destroy Néel order.[2]

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