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Spin correlations in a two-dimensional Heisenberg antiferromagnet doped with static holes

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We study the temperature dependence of the spin correlation length of the spin-$\frac{1}{2}$ Heisenberg antiferromagnet doped with static holes which are randomly distributed in the square lattice. We have used Handscomb's quantum Monte Carlo method to study lattices of sizes up to 20×20 and the high-temperature series expansion. In the doping regime where the system is ordered at $T = 0$, the correlation length can be fit at low temperature by the form $\xi(T) = c(x) \exp[2\pi\rho_s(x)/T]$, where the dependence of the spin stiffness constant $\rho(x)$ and of the prefactor $c(x)$ on doping x can be understood by simple scaling arguments. We discuss the implications of these results on the neutron-scattering data obtained on lightly doped copper oxides.

One of the most intensively studied aspects of the copper oxide superconductors is their magnetic properties. In particular, since these materials are believed to be strongly correlated electronic systems, and their superconducting phase is near their antiferromagnetic and metal-insulator instability there is a school of thought[1] that believes that perhaps these phenomena are intimately related.

Among the quantities providing very illuminating information about the dynamics of the spin degrees of freedom in the materials is the temperature dependence of the spin-dynamical structure function as seen by neutron-scattering experiments.[2] The temperature dependence of the correlation length in the pure La_2CuO_4 can be well described by the spin-$\frac{1}{2}$ antiferromagnet on the square lattice[3] using a value of the antiferromagnetic coupling $J \simeq 1500$ K. The correlation length in the undoped spin-$\frac{1}{2}$ antiferromagnet at low temperatures increases very rapidly following the exponential form

$$\xi(T) = c \exp(2\pi\rho_s/T), \tag{1}$$

where $\rho_s = 0.20$ J, and it is the spin stiffness constant of the spin-$\frac{1}{2}$ antiferromagnet on the square lattice and $c = 0.276a$ (Ref. 3), where a is the distance between nearest-neighboring Cu spins ($a \simeq 3.8$ Å). This form was predicted by Chakravarty, Halperin, and Nelson[4] by mapping the model to the quantum-mechanical nonlinear σ model in two-space dimensions and one Euclidean time dimension of length $\hbar c\beta$. This behavior has been demonstrated to be valid at low temperatures by quantum Monte Carlo simulations.[3]

There are neutron-scattering data[5] for the behavior of the correlation length as a function of temperature for lightly doped $La_{2-x}Sr_xCuO_4$ for small x (where the material is believed to be in a spin-glass phase). In the low doping limit, it is believed that holes are static because in this limit the material behaves as an insulator; thus the mass of the holes is large enough to localize the holes around the Sr atoms due to the Coulomb attraction.

In this paper we simulate the quantum spin-$\frac{1}{2}$ antiferromagnetic Heisenberg model (AHM) doped with static holes, uniformly distributed on the square lattice, using Handscomb's quantum Monte Carlo method for antiferromagnets.[3] The calculation is performed for various hole concentrations, temperatures, and size lattices. We find that, in the low doping regime when the system is ordered at $T = 0$, the correlation length behaves as given by Eq. (1) with spin-stiffness constant $\rho_s(x)$ and the prefactor $c(x)$ both functions of the doping fraction x. The dependences of $\rho_s(x)$ and $c(x)$ on x are those that one predicts based on simple scaling arguments with x. Our results are compared with the experimental results and we discuss possible reasons for the disagreement found at low temperatures.

Let us consider the spin-$\frac{1}{2}$ AHM doped with static holes, which are randomly distributed on the square lattice of size $N = L \times L$:

$$H = J \sum_{<ij>, i,j \neq \{l_k\}} \mathbf{S}_i . \mathbf{S}_j , \tag{2}$$

where the sum is over all nearest-neighbor pairs $< ij >$ of spins in the CuO_2 plane, which do not contain holes. The holes are located at sites $l_1, l_2, \ldots, l_n$ and the doping fraction is $x = n/N$. Here $\mathbf{S}_i$ is the spin-$\frac{1}{2}$ operator of the conduction-band electron located at the ith cell.

The Hamiltonian (2) apart from a constant can be written as

$$H = -J/2 \sum_{<ij>, i,j \neq \{l_k\}} (h_{ij}^2 - h_{ij}) + \text{const.} \tag{3}$$

The operator h_{ij}, when there is a hole at the site i or j, gives zero, and when there is no hole at either i or j, the operator h_{ij} is equal to $S_i^+ S_j^- + S_i^- S_j^+$, i.e., flips antiparallel spins and gives zero in the case of parallel spins.

Using Handscomb's Monte Carlo method an observable quantity can be calculated by expanding $e^{-\beta H}$ as

an infinite sum of sequences of bond operators h_{ij} or h_{ij}^2. Then one performs sampling in an abstract space of sequences $C_r = \{i_1, i_2, ..., i_r\}$ of $r = r_1 + r_2$ operators, where r_1 (r_2) is the number of h's (h^2's) in the sequence. The expectation value of an observable quantity is calculated as an average over a distribution of sequences. This method for the case of the pure Heisenberg antiferromagnet has been outlined in detail elsewhere including in Ref. 3. In general, one needs to define a Markov chain generating the correct distribution $\pi(C_r)$ of sequences C_r. In the present case some of the bonds are broken due to the presence of holes. Let the number of unbroken bonds be N_b. At each iteration of the random walk we can add or remove any number n_a or n_d of operators respectively from a total of $2N_b$ operators. We decide to add or delete an operator with equal probability. We select a given operator to be added with probability $\frac{1}{2N_b}$ and the specific location in a string with r operators with probability $\frac{1}{r+1}$. We remove a given operator with probability $\frac{1}{r}$. The acceptance probability for a transition from the state C_r having r operators to the state $C'_{r'}$ having $r' = r + n_a - n_d$ operators and satisfying the detailed balance is given by

$$P(C_r \to C'_{r'}) = \min\left[1, w\left(\frac{1}{2N_b}\right)^{r-r'} \frac{\pi(C'_{r'})}{\pi(C_r)}\right], \quad (4)$$

where the factor $w = \frac{1}{2}$, 2, or 1, for $r = 0$, $r' = 0$, or $r \neq 0$ and $r' \neq 0$, respectively.

We have performed calculations on lattices with sizes 10^2 and 20^2 with periodic boundary conditions. The number of iterations we performed depended on the temperature and lattice size and, in general, we generated on the order of $10^6 - 10^7$ iterations for thermalization, and on the order of 10^7 iterations for measurements. The quantities that required more iterations to reach equilibrium were r_1 and the spin correlation function. Simulations with this algorithm at lower temperatures than the ones reported here or larger lattices are beyond realistic computational time scales.

We calculated the staggered correlation function $G(r)$ defined as follows:

$$G(\mathbf{r}) \equiv (-1)^{|x+y|} \frac{4}{L^2} \sum_n < S_n^z S_{n+\mathbf{r}}^z >, \quad (5)$$

where the summation is over all occupied pairs n and $n+\mathbf{r}$. We fit G to the form $\lim_{r\to\infty} G(\mathbf{r}) = A\cosh(\frac{\mathbf{r}-L/2}{\xi(T)})$. Figure 1 shows G calculated for the 10×10 and 20×20 lattices for $T = J$ and $x = 0$ and $x = 0.2$. The solid and dashed lines are obtained by fitting all except the first few points of G to a cosh. The data fitted by the dashed line correspond to $x = 0$ [giving $\xi(T = J, x = 0) = 0.88 \pm 0.03$], while those fitted by the solid line correspond to $x = 0.2$ [giving $\xi(T = J, x = 0.2) = 0.70 \pm 0.04$].

In Table I and Fig. 2 we give the results for the inverse correlation length as extracted from our calculation. We have omitted the error bars from the figure for clarity. Notice that for $T > 0.5$ J the size dependence of the correlation length for lattices 10×10 and 20×20 is negligible as has also been noted before.[3] For lower temperatures,

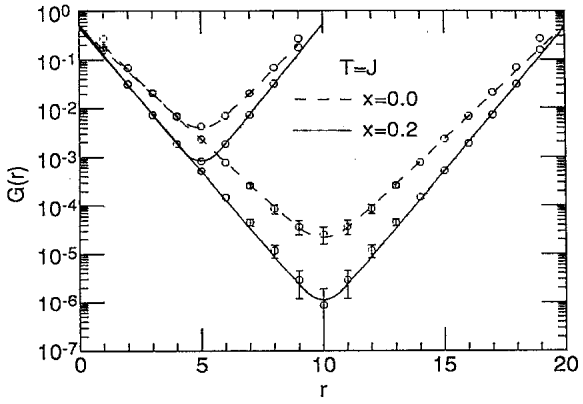

FIG. 1. The staggered correlation function calculated for the 10×10 and 20×20 lattices for $T = J$ and for $x = 0$ and $x = 0.2$. The solid ($x = 0.2$) and dashed lines ($x = 0$) are obtained by fitting all except the first few points of G to a cosh.

in order to avoid finite-size effects, one needs to study larger size lattices; the spin relaxation time, however, becomes extremely large, and our method becomes very inefficient.

The leading-order contribution to the correlation function $G(r)$ in high-temperature series expansion can be calculated as in Ref. 3. The leading contribution to $G(r)$ is of rth order in βJ because we need to introduce at least r operators between the spin at n and the spin at $n + r$ to obtain a connected diagram. We find

$$\lim_{\beta J \to 0} G(r) = (1 - x)^r \left(\frac{\beta J}{4}\right)^r. \quad (6)$$

The factor $(\beta J/2)^r$ comes from the order of the expansion, and the calculation of the trace gives 2^{-r}, while the factor $(1 - x)^r$ gives the probability that there is no

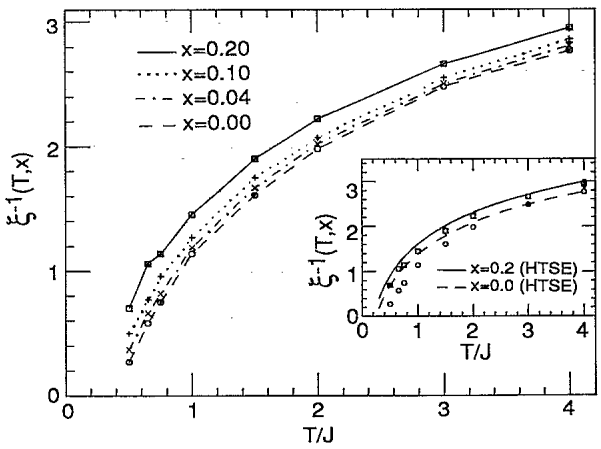

FIG. 2. Our results for the inverse correlation length as a function of T for doping fractions $x = 0, 0.04, 0.10, 0.20$. In the inset we compare the results of the quantum Monte Carlo calculation given by the open circles and squares with the results of the high-temperature series expansion (HTSE) given by the dashed and solid lines for $x = 0$ and 0.2, respectively.

TABLE I. The inverse correlation length $\xi^{-1}(T,x)$ of the doped and undoped system as a function of temperature for doping ratios $x = 0.04, 0.1$, and 0.2.

	$x = 0.0$		$x = 0.04$		$x = 0.1$		$x = 0.2$	
T	10×10	20×20	10×10	20×20	10×10	20×20	10×10	20×20
0.5	0.27(1)		0.37(1)		0.50(1)		0.70(1)	
0.65	0.58(2)	0.58(3)	0.66(4)	0.63(3)	0.77(1)		1.06(3)	
0.75	0.75(2)		0.82(1)		0.96(3)		1.14(1)	
1.0	1.14(3)	1.12(3)	1.19(2)	1.20(3)	1.27(1)	1.26(5)	1.45(1)	1.40(5)
1.5	1.61(2)		1.67(2)		1.75(2)		1.90(1)	
2.0	1.98(1)	1.98(3)	2.02(3)	2.02(3)	2.07(2)	2.11(3)	2.22(1)	2.22(5)
3.0	2.48(2)		2.50(3)		2.55(5)		2.66(3)	
4.0	2.77(3)		2.81(3)		2.86(3)		2.95(8)	

hole occupying the r sites between n and $n+r$. If there is a hole at one of the sites of the shortest path joining the sites n and $n+r$, we need to choose a longer path of bonds in order to create a connected diagram. This contribution will be of order $(\beta J)^{r+2}$ or higher. Thus $G(r) = \exp[-r/\xi(T)]$ and

$$\lim_{T\to\infty} \xi^{-1}(T,x) = ln(4T/J) - ln(1-x). \tag{7}$$

This expression is compared with the numerical results in the inset of Fig. 2 for $x = 0$ and 0.2. Notice that, while the agreement is good at high temperatures, at $T/J < 1$ the agreement becomes worse.

We have analyzed our numerical results by making the following hypothesis: in the region of x in which the model is ordered at $T = 0$, the model at *long wavelengths* should behave in the same way as the undoped antiferromagnet with a modified spin-stiffness constant. Namely, the system can be described by a nonlinear σ model with a reduced value of the spin-stiffness constant, thus getting closer to its quantum critical point, which separates the ordered phase from the quantum disordered phase.

In Fig. 3, we show that the correlation length at various values of x as a function of T follows similar exponential behavior as the undoped system ($x = 0$):

$$\xi(T,x) = c(x)\exp(2\pi\rho_s(x)/T) \tag{8}$$

by plotting $-T\ \ln[\xi(T,x)]$ as a function of T. Here the straight lines are fits giving values for the spin-stiffness constants plotted as a function of x in the inset of Fig. 3 (open circles). The constant prefactor $c(x)$, which is obtained from the fit, is also shown in the inset of Fig. 3 (open squares). Notice that the quality of the fit for $x \neq 0$ to the form (8) is as good as the $x = 0$ case (open circles and dashed line), which is believed to follow such a behavior.[4,3]

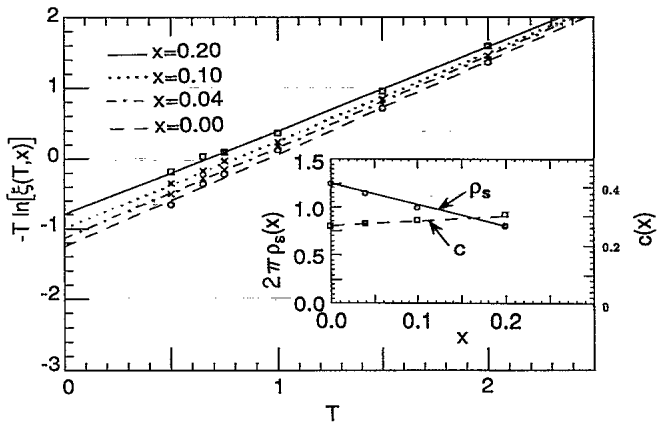

FIG. 3. We demonstrate that the results of our Monte Carlo calculation for the correlation length follow the exponential form given by Eq. (8) at low T. In the inset the open circles and squares give the values of $\rho_s(x)$ and $c(x)$ found from the fit to Eq. (8), while the solid and dashed lines give the functions (9) and (10) predicted by the simple scaling argument.

The spin-stiffness constant at a finite doping x should behave approximately as

$$\rho_s(x) = \rho_s(0)(1-x)^2 \tag{9}$$

because the density of unbroken antiferromagnetic bonds is reduced (due to the presence of the holes) by a factor $(1-x)^2$. Notice that the approximate Eq. (9) with $\rho_s(0) = 0.20$ J, shown by the solid line in the inset of Fig. 3, approximates very well the values of $\rho_s(x)$ found from the fit. The constant $c(0)$ in the undoped system is given by $0.276a$, where a is distance between two nearest-neighboring spins. The average distance between two nearest-neighboring spins has been increased in the doped system by a factor $1/\sqrt{1-x}$; thus we expect that

$$c(x) = c(0)/\sqrt{1-x}, \tag{10}$$

which is shown by the dashed line in the inset of Fig. 3 and approximates very well the results of our calculation.

Combining the above three equations we find that if we rescale the temperature to $T' = T/[(1-x)^2]$ and the correlation length to $\xi' = \xi\sqrt{1-x}$ all the results should approximately collapse onto the same curve. Namely, ξ' versus T' is given by the $x = 0$ curve [Eq. (1)]. In Fig. 4 we demonstrate that the correlation length as a function of temperature and for $x = 0, 0.04, 0.1, 0.2$ rescaled as predicted above collapses onto the same curve. The solid line is the same curve as that given by Eq. (1), and it has been obtained by fitting our $x = 0$ results at low temperatures. There is some disagreement between the solid line and the numerical results at high temperatures as expected [the form (1) is only valid at low T/J]. Clearly this and the fact that $\rho_s(x < 0.2) \neq 0$ indicates

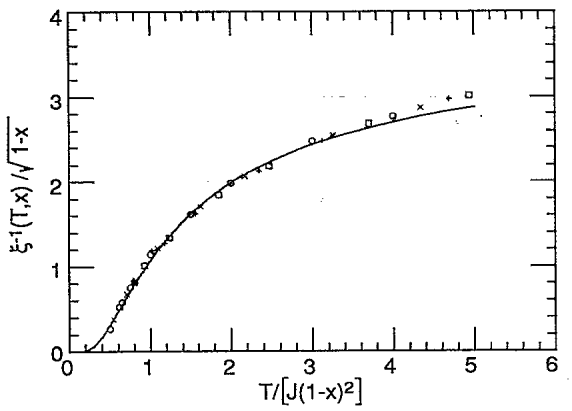

FIG. 4. We demonstrate that the temperature dependence of the correlation length for various values of the doping ratio x (we have used different symbols in the plot for different values of x) and as a function of temperature collapse onto the same curve by scaling the temperature and the lengths by simple functions of x. The solid line is the exponential form (1), which has been obtained by fitting the $x = 0$ results at low temperatures.

that the critical point separating the ordered phase from the quantum disordered phase at $T = 0$ is $x_c > 0.2$.

We can compare the neutron-scattering data for $\xi^{-1}(T, x)$ obtained on the lightly doped quantum antiferromagnet $La_{1.96}Sr_{0.04}CuO_4$ with those obtained from the present calculation for $x = 0.04$ by extrapolation to low temperatures using the form (9). We find that the agreement is reasonable for high $T > 450$ K. However, at low T ($T < 300$ K) the experimentally measured correlation length is temperature independent, while the theoretical curve for $\xi(T, x = 0.04)$ is strongly temperature dependent. There could be a number of reasons for the disagreement at low T. First, our extrapolation formula (8) may not be valid at low T, where holes or other impurities may cause the system to be in a spin-glass phase. We would like to emphasize that temperatures less than 0.5 J are not accessible to our calculation because the spin relaxation time, measured in terms of the number of iterations required for thermalization of a given starting spin configuration, is very large. Thus, it is possible that the Heisenberg antiferromagnet doped with static holes can be described by such a spin-glass phase at low temperatures. A second possible reason for the disagreement could be that the holes in the actual system are mobile, even in this low doping case, and thus create more damage to the spin background and to the correlations between spins. Monte Carlo studies of models with finite density of mobile holes (such as the t-J model) on large size lattices (needed in order to accurately calculate the correlation length) are hindered by the fermion sign problem. A third possibility is that the holes are indeed static, but they change the sign of the spin-spin interaction in their neighborhood,[6] and this creates more dramatic effects in the behavior of the spin-spin correlation length. The last possibility can be explored by studying the antiferromagnetic Heisenberg model on the square lattice, where we introduce a finite density of ferromagnetic bonds.

Note added in proof. There are measurements of the temperature dependence of the correlation length $\xi(T, x)$ for electron-doped copper oxides such as $Nd_{2-x}Ce_xCuO_4$ by Matsuda *et al.*[7] In this material the behavior of $\xi(T, x)$ is qualitatively similar to that given by our Eq. (8) and qualitatively different from that in $La_{2-x}Sr_xCuO_4$. The difference between these two systems favors the point of view that the amount of local frustration (such as the third possibility discussed above) is much greater in the hole-doped materials as compared to that in the electron-doped materials. Thus, our model is more appropriate to describe the spin correlations in the electron-doped materials in the low-x limit, where the carriers are localized.

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