Magnetic Susceptibility Scaling in $La_{2-x}Sr_xCuO_{4-y}$

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The powder magnetic susceptibility $\chi(T)$ of $La_{2-x}Sr_xCuO_{4-y}$ is found to scale with doped hole concentration $p=x-2y$ according to a law of corresponding states for $0\leq p\leq 0.20$, thereby allowing $\chi^{\text{Pauli}}(p)$ of the holes and $\chi^{2D}(p,T)$ of the Cu^{+2} spin sublattice to be separated and precisely evaluated. χ^{Pauli} increases with p. The shape of $\chi^{2D}(T)$ is that of the spin-$\frac{1}{2}$ square-lattice Heisenberg antiferromagnet; however, by $p=0.20$, the in-plane Cu-Cu superexchange coupling constant and effective magnetic moment per Cu ion are both largely suppressed.

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Evidence has been accumulating that superconductivity in the high-T_c copper oxides develops in the presence of dynamic two-dimensional short-range antiferromagnetic ordering of the Cu^{+2} spin sublattice.[1-5] This has important implications for the superconducting mechanism. Herein, we report that the powder magnetic susceptibility $\chi(T)$ of $La_{2-x}Sr_xCuO_{4-y}$ scales with doped hole concentration $p=x-2y$ according to a law of corresponding states, which bears directly on this issue. From the scaling parameters, we have separated and precisely determined the effective susceptibilities $\chi^{\text{Pauli}}(p)$ of the doped hole carriers and $\chi^{2D}(p,T)$ of the Cu^{+2} spin sublattice for the first time. These will be derived and discussed.

$\chi(T)$ data[2,3] for five $La_{2-x}Sr_xCuO_{4-y}$ samples are shown in Fig. 1. Curie (C/T) terms, corresponding respectively to $\simeq$0.28% and 0.57% (gyromagnetic factor $g=2$) of the Cu^{+2} spins residing as isolated defects or in impurity phases, have been subtracted for $x=0.05$ and 0.2(II); the correction for 0.2(II) was determined from the analysis below. For $x=0$, only the data above the structural transition at $T_0=530$ K have been included, because below this T ferromagnetic correlations appear and grow with decreasing T,[6] strongly affecting $\chi(T)$.[2,3,6] The $x=0.2$ (I and II) data show smooth broad maxima at $T\equiv T^{\max}(p)$, where $\chi\equiv\chi_{\max}(p)$. Superconductivity data for the samples are listed in Table I. The data in Fig. 1 are very similar to those in Ref. 7. The evolution of $\chi(T)$ with O doping in the $YBa_2Cu_3O_{6+x}$ system[2,3] is similar to that in Fig. 1.

Inspection of Fig. 1 suggests that $\chi(T)$ scales simply with p:

$$\chi(p,T)=\chi_0(p)+[\chi_{\max}(p)-\chi_0(p)]F(T/T^{\max}(p)), \quad (1)$$

where $\chi_0(p)$ is independent of T and $F(z)$ is a universal function. The T dependence of χ is attributed to the effective susceptibility $\chi^{2D}(p,T)$ of the Cu^{+2} spin sublattice, which possibly includes renormalization via interaction with doped hole spins (see below), so that $\chi^{2D}(T)=\chi(T)-\chi_0$ and $\chi^{2D}_{\max}=\chi_{\max}-\chi_0$. Nonlinear regression analyses yielded the scaling parameters $T^{\max}(p)$ and $\chi_{\max}(p)$, listed in Table I, and gave $\chi_0(p)$ to within a constant additive factor. This contribution is taken to be

$$\chi_0(p)=\chi^{\text{core}}+\chi^{VV}+\chi^{\text{Pauli}}(p). \quad (2)$$

From standard tables, the isotropic atomic core susceptibility is $\chi^{\text{core}}=-9.9\times10^{-5}$ cm^3/mol. A powder average of the anisotropic T-independent Van Vleck contribution for Sc_2CuO_4 (Ref. 8) gives $\chi^{VV}=+2.4\times10^{-5}$ cm^3/mol. χ^{core} and χ^{VV} are taken to be independent of composition over the limited x and y ranges studied here. Assuming $\chi^{\text{Pauli}}(0)\equiv0$ fixes the above additive constant and yields the $\chi_0(p)$, $\chi^{\text{Pauli}}(p)$, and $\chi^{2D}_{\max}(p)$ values in Table I. Possible contributions to χ from Landau diamagnetism are absorbed into χ^{Pauli}. The derived $F(T/T^{\max})=\chi^{2D}(T/T^{\max})/\chi^{2D}_{\max}$ is plotted versus $T/T^{\max}$ in Fig. 2. The solid curves are the data for the $x=0.2$(I) sample; for clarity only a few representative points from each of the other data sets are shown. It is clear from Fig. 2 that the $\chi^{2D}(T/T^{\max})/\chi^{2D}_{\max}$ data lie on a common curve and that this curve parametrizes the shape of $\chi(T)$ for each of the samples well.

We find $p=x$ for the $x=0$, 0.05, 0.1, and 0.2(II) samples, but $p<x$ for the $x=0.2$(I) sample. For the first

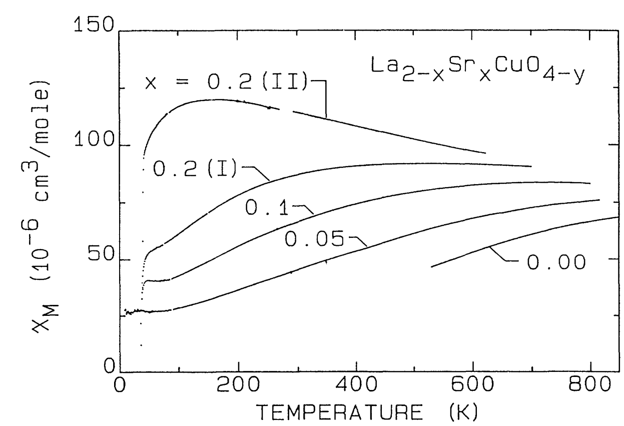

FIG. 1. Magnetic susceptibility χ vs temperature for samples of $La_{2-x}Sr_xCuO_{4-y}$ (Refs. 2 and 3).

TABLE I. Parameters for samples of $La_{2-x}Sr_xCuO_{4-y}$. The Meissner effect (ME) was measured in a magnetic field of 50 G, and the superconducting transition temperature T_c is the midpoint of the ME transition.

Composition[a,b] x	y	p	T_0 (K)[a,b]	ME (%)[b]	T_c (K)[b]	T^{max} (K)	χ_{max}[e]	χ_0[e]	χ^{Pauli}[e]	χ^{2D}_{max}[e]
0	0.043	0[a]	530	0	⋯	1460	7.93	≡ −7.5	≡0	15.4
0.05	0	0.05	415	0	⋯	1340	8.15	−1.49	5.01	9.64
0.1	0	0.1	290	6	22	823	8.39	0.38	7.88	8.01
0.2(I)	0.04[c]	0.13	>290	30	31	515	9.17	1.94	9.44	7.23
0.2(II)	0	0.2	<24	16[d]	23[d]	170	11.97	6.48	14.0	5.49

[a]Reference 2.
[b]Reference 3.
[c]Our scaling results give $y = 0.035 \pm 0.005$.
[d]The reason that this apparent T_c is lower than in Fig. 3 for $p = 0.2$ and that the ME fraction is low is unknown; it may be related to strong flux pinning in this particular sample.
[e]10^{-5} cm^3/mol.

four samples, $p = x$ is inferred from the manner in which the samples were prepared[2,3] and from a comparison of the observed[2,3] $T_0(x)$ values (Table I) with those in Ref. 12. The T_0 values for the samples with $x \leq 0.1$ are well defined ($\simeq \pm 10$ K); since $dT_0/dp \simeq -2700$ K,[12] the maximum inhomogeneity in p within the samples is estimated to be $\simeq \pm 0.004$, except possibly for the $x = 0.2$(I) sample which was synthesized in pellet form rather than as a powder. The y values (± 0.02) from thermogravimetric analyses are listed in Table I.[2,3] From the scaling parameters in Table I for the first four samples, $p = 0.13 \pm 0.01$ is inferred for the $x = 0.2$(I) sample; this value is consistent with the T_0 value and O stoichiometry since $p = x - 2y$. By comparing $T_0(x = 0.2(\text{I}), p = 0.13)$ with $T_0(x = 0.2(\text{II}), p = 0.2)$, *the primary parameter determining* T_0 *is not x but p.*[2]

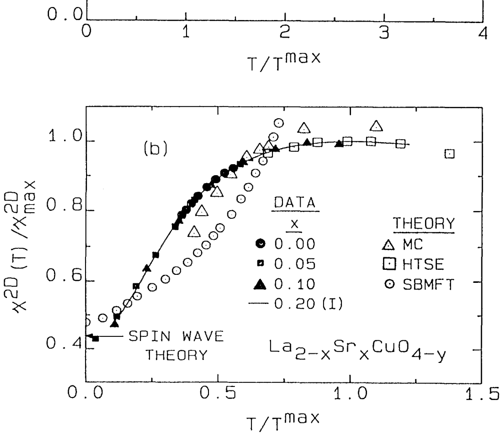

FIG. 2. Cu^{+2} sublattice susceptibility $\chi^{2D}(T)/\chi^{2D}_{max}$ vs T/T^{max} for the samples in Fig. 1. Theoretical predictions (Refs. 9–11) and the Weiss molecular-field (MFT) prediction are shown for comparison.

We now discuss $\chi^{Pauli}(p)$ and return to $\chi^{2D}(p,T)$ below. χ^{Pauli} increases with p, as seen in Table I. From our perspective, the existence of χ^{Pauli} implies (i) the existence of a Fermi energy E_F in a continuous energy distribution of fermion-doped quasihole states, (ii) degeneracy of spin-up and spin-down quasihole states near E_F, and (iii) accessibility of the empty states near E_F to quasiholes at or near E_F with appropriate excitation (magnetic field and/or temperature). For insulators like the $p = 0.05$ sample with $\chi^{Pauli} > 0$ (see below), feature (iii) implies (iv) that the quasihole localization length is sufficiently large that the density of states at E_F can be effectively sampled. These four conditions are also sufficient to predict that a Sommerfeld heat capacity (C_p) coefficient $\gamma \propto \chi^{Pauli}$ should be observed at low T for insulating compositions. $\chi^{Pauli}(p)$ in Table I should provide a testing ground for theoretically predicted quasihole Fermi surfaces.

The magnetic-susceptibility density of states at E_F $[D_\chi(E_F)]$ is

$$D_\chi(E_F) = \chi^{Pauli}/\mu_B^2 , \tag{3}$$

which assumes $g = 2$ for the doped quasihole spins. For $p = 0.2$, $D_\chi(E_F) = 4.3$ states/eV-f.u. (where f.u. stands for formula unit). Comparison of this value with the bare $D(E_F)$ (2.1 states/eV-f.u.) from (single particle) band-structure calculations[13] suggests a Stoner enhancement factor $S = [1 - ID(E_F)]^{-1} \simeq 2.0$. This S is close to the value 1.7 calculated[14] using the band-theory value for the exchange correlation integral $I = 0.20$ eV-f.u. in

Sc_2CuO_4[8] and the bare $D(E_F)$ for $p=0.2$ above. This agreement suggests that whereas band theory fails to predict the observed properties of La_2CuO_4, band theory may be more reliable for the heavily doped compositions.

Significantly, the doped quasiholes in the insulating[15] $p=0.05$ sample give rise to a paramagnetic T-independent χ contribution (χ^{Pauli}) rather than diamagnetism or a Curie law. As noted above, a nonzero γ should therefore be observed at low T for $p=0.05$. In the absence of theory for γ in the cuprates, we test the degenerate Fermi-*gas* prediction,

$$\gamma=(\pi^2/3)k_B^2 D_\chi(E_F)\,. \tag{4}$$

Equations (3) and (4) predict $\gamma(0.05)=3.7$ mJ/mol-K^2 using $\chi^{\text{Pauli}}(0.05)$ in Table I. This γ value is in surprisingly excellent agreement with $\gamma=3.9\pm0.3$ mJ/mole-K^2 observed[15] in the *low-T* C_p for $p=0.05$, confirming (4). Given this agreement, we use (3), (4), and Table I to predict *normal-state* γ values of 6.9 and 10.2 mJ/mol-K^2 for the superconducting compositions with $p=0.13$ and 0.20, respectively. A lower limit on the C_p discontinuity at T_c for $La_{1.85}Sr_{0.15}CuO_4$ is $\Delta C_p/T_c=20\pm5$ mJ/mol-K^2.[16] Our predicted $\gamma(0.15)\simeq8$ mJ/mol-K^2 then yields $\Delta C_p/\gamma T_c \gtrsim 2$, indicating that this compound is a strong-coupling superconductor, consistent with Ref. 16.

We return now to the $\chi^{2D}(p,T)$ data in Fig. 2. Within the range $T/T^{\max}=0.69$ to 4.0 over which the accurate high-T series expansion (HTSE) calculations[9] for the spin-$\frac{1}{2}$ square-lattice Heisenberg antiferromagnet were reported, the data in Fig. 2 are consistent with this prediction, as shown. As $T\rightarrow0$, the data closely approach the spin-wave-theory value.[10] In the theoretically difficult intermediate-T range [Fig. 2(b)], our results are in better agreement with Monte Carlo (MC) results for a 16×16 square lattice[11] than with a Schwinger boson mean-field-theory (SBMFT) prediction,[10] except near $T=0$.

The data in Fig. 2 are therefore consistent with a picture in which localized spins are present on the Cu^{+2} ions in $La_{2-x}Sr_xCuO_{4-y}$ and exhibit dynamic Heisenberg antiferromagnetic intralayer order throughout the metallic as well as the insulating composition regimes for $0\leq x\leq0.20$, consistent with Refs. 1-5. That the (effective) magnetic behaviors of the doped holes and of the Cu spin layers can apparently be separated in this strongly interacting many-body system is significant; this result was anticipated theoretically.[17] Hole doping does have two important influences on the Cu sublattice magnetism as shown below: (i) It strongly reduces the effective intralayer Cu-Cu superexchange coupling constant J and (ii) it strongly reduces the effective moment per Cu ion.

Theoretically, for the spin-$\frac{1}{2}$ square-lattice Heisenberg antiferromagnet, $J=T^{\max}/1.86$.[9] From this relation and Table I, $J(p)$ was computed and is plotted in

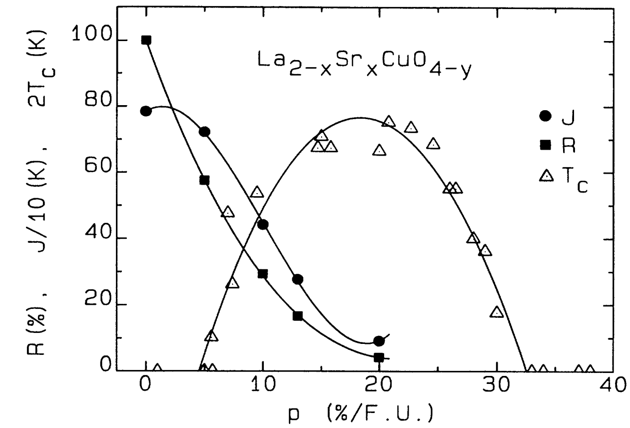

FIG. 3. Effective intralayer Cu-Cu exchange coupling constant J and the ratio $R\equiv\chi_{\max}^{2D}/\chi_{\max}^{2D\,\text{calc}}$ vs p. $T_c(p)$ (Ref. 18) is for comparison. The solid curves are guides to the eye.

Fig. 3, where J is seen to decrease strongly with p to a value of 91 K at $p=0.20$. The value of $J(0)=780$ K is in agreement with values from Ref. 1. The rapid drop in J with p has been noted previously.[2,4,5] This result is also supported by recent inelastic neutron scattering experiments on single crystals of $La_{2-x}Ba_xCuO_{4-y}$, which show that the spin-wave velocity decreases noticably with p.[19] If the doped holes go on O ions in the CuO_2 layers, the superexchange coupling between the Cu ions adjacent to these O ions would be mostly destroyed,[17] thereby reducing the effective J.

On the other hand, from the HTSE results,[9] $\chi_{\max}^{2D\,\text{calc}}=g^2(0.0176\ \text{cm}^3\text{-K/mole})/J$. Thus, if J decreases, the observed $\chi_{\max}^{2D}$ should increase proportionately. This is found *not* to be the case, notwithstanding Fig. 2. The ratio $R\equiv\chi_{\max}^{2D}/\chi_{\max}^{2D\,\text{calc}}$ is plotted versus p in Fig. 3. R decreases rapidly from 1 at $p=0$ ($g=2.62$) to 0.041 at $p=0.20$, assuming that g is independent of p. Inclusive of possible changes in g with p, this decrease corresponds to a decrease in the effective moment per Cu ion by a factor of 5.0. A possible explanation is that doped hole spins on the oxygen ions form nonmagnetic singlet states with adjacent Cu^{+2} spins[20]; dopant-enhanced d^9-d^{10} Cu valence fluctuations[17] would also reduce the effective moment.

The implications of these results for the superconducting mechanism are not yet clear. $T_c(p)$ (Ref. 18) is plotted in Fig. 3, where strong anticorrelations between J (and R) and T_c are seen; the former was first noted by Aeppli.[4] At first sight, these suggest that antiferromagnetic correlations suppress superconductivity rather than serve as a vehicle[21] for its occurrence. However, other interpretations are also plausible. We point out that although J and R for $p=0.2$ in Fig. 3 are small relative to their values near $p=0$, χ^{2D} still comprises, e.g., $\simeq40\%$ of the observed value in Fig. 1 (cf. Table I).

In conclusion, when quantitative predictions of $\chi(p,T)$

for the various proposed superconducting pairing models become available, our results should help to determine whether the mechanism is magnetic in origin and to perhaps identify the specific one(s) operational in the high-T_c cuprates.

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