Antiferromagnetic exchange in two-leg spin-$\frac{1}{2}$ ladders

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Extraction of antiferromagnetic (AF) exchange constants J and J' in two-leg spin-$\frac{1}{2}$ nearest-neighbor (NN) Heisenberg ladders from experimental magnetic spin susceptibility $\chi(T)$ data is studied, where J' is the NN exchange constant in the rungs and J is that in the legs. Two low-T ($T \ll J/k_B$) approximations of Troyer, Tsunetsugu, and Würtz [Phys. Rev. B **50**, 13 515 (1994)] for $\chi(T)$ of the isolated spin-$\frac{1}{2}$ two-leg ladder with $J'/J=1$ are shown to be accurate to $\sim 10\%$ when extrapolated to $T \sim J/k_B$. The variations in the magnetic excitation dispersion relation parameters [which enter the low-T expression of Troyer *et al.* for $\chi(T)$] for $J'/J<1$ are inferred from previous theoretical results. Next, the values of the maxima in $\chi(T) \equiv \chi^{\max}$ from many numerical calculations in the literature for various nonfrustrated spin-$\frac{1}{2}$ NN Heisenberg antiferromagnets are considered. The values of the product $\chi^{\max} z_{\rm eff}$ are found to depend systematically and almost uniqely on an effective magnetic coordination number $z_{\rm eff}$, defined in the text, but are nearly independent of the dimensionality of the spin lattice and of the magnitude of any gap in the magnetic excitation spectrum. This observation allows accurate bounds on the exchange coupling constants in arbitrary quasi-low-dimensional spin-$\frac{1}{2}$ antiferromagnets to be inferred from experimental $\chi^{\max}$ values. Finally, a mean-field-type expression for $\chi(T)$ for arbitrary quasi-low-dimensional spin-$\frac{1}{2}$ Heisenberg antiferromagnets is derived, using the above $\chi^{\max}$ phenomenology as input, which allows the influence of intersystem coupling on $\chi(T)$ to be evaluated. These results, and the $\chi(T)$ calculations of Barnes and Riera [Phys. Rev. B **50**, 6817 (1994)] for isolated two-leg spin-$\frac{1}{2}$ Heisenberg ladders, are used to analyze the experimental $\chi(T)$ data of Azuma *et al.* [Phys. Rev. Lett. **73**, 3463 (1994)] for the spin-$\frac{1}{2}$ two-leg ladder compound $SrCu_2O_3$. The analyses together suggest that $J'/J \sim 0.5$, contrary to the expectation that $J'/J \approx 1$, and that J is very large (~ 2000 K), similar to the value of J in the linear-chain cuprate Sr_2CuO_3. [S0163-1829(96)01442-7]

I. INTRODUCTION

Spin configurations formed by coupling n spin chains side by side in a plane are termed n-leg spin ladders. The study of spin-$\frac{1}{2}$ ladders with increasing numbers of legs is one way to approach the physics of the square lattice of spins as in the layered cuprate superconductor parent compounds, corresponding to $n \to \infty$. Such spin ladders have received increasing attention over the last several years.[1] For antiferromagnetic (AF) Heisenberg exchange interactions between nearest-neighbor (NN) spins, the only case discussed here, the spin Hamiltonian of the n-leg spin ladder is

$$\mathcal{H} = J \sum_{\langle i,j \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_j + J' \sum_{\langle i,k \rangle} \boldsymbol{S}_i \cdot \boldsymbol{S}_k , \qquad (1)$$

where $J, J' \geq 0$ for AF coupling. The first sum is over distinct NN spin pairs in each chain (over legs of the ladder), and the second is over distinct NN spin pairs in adjacent chains (over rungs of the ladder). The spin exchange coupling constant is J within a leg and J' within a rung.

Experimental research on spin ladders was stimulated by theoretical predictions that the (unfrustrated) spin-$\frac{1}{2}$ two-leg ladder should have a nonmagnetic quantum ''spin-liquid'' ground state, with a spin-gap Δ to the lowest magnetic triplet excited states,[2–4] in contrast to the isolated chain which has no spin gap. This prediction was verified for the spin-$\frac{1}{2}$ two-leg ladder compounds $(VO)_2P_2O_7$ (Refs. 5 and 6) and $SrCu_2O_3$.[7–12] The ground state of $SrCu_2O_3$ is evidently very close to the critical point separating the antiferromagnetically ordered state from the spin liquid, since slightly perturbing the system by replacing only $\sim$ 1% of the Cu by Zn is sufficient to induce long-range AF order.[13] The two-leg ladder compound $LaCuO_{2.5}$ (high pressure form) was initially reported to have a spin-liquid ground state with a spin gap,[14] but was subsequently inferred to order magnetically at ~ 110 K.[15] Calculations indicate that the ratio of the interladder to intraladder exchange coupling constants in this compound is $\sim$ 0.1 to 0.25.[16,17]

Barnes and Riera[6] calculated the spin susceptibility versus temperature $\chi(T)$ for two-leg spin-$\frac{1}{2}$ ladders and obtained an excellent fit with $J \approx J'$ to the $\chi(T)$ data[5] for $(VO)_2P_2O_7$. They also predicted that $\Delta \approx J/2$ (at wave vector $k=\pi/a$ along the ladder where a is the spin-spin distance) for this compound, which was subsequently confirmed by neutron scattering measurements.[18] For wider ladders, the (unfrustrated) even-leg ladders are predicted to continue to show spin gaps with a magnitude decreasing with n, whereas the odd-leg ladders display behavior similar to that of the gapless isolated linear chain.[1,8,19–22] The latter prediction was verified for the three-leg ladder compound $Sr_2Cu_3O_5$, which showed no spin gap[7–10] and exhibited disordered static AF ordering below ~ 50 K.[11] Also stimulating the experiments on such materials were predictions that superconductivity might occur by a purely electronic mechanism in weakly coupled and weakly doped even-leg ladders.[1,3,8,23,24] To date, the only spin-ladder compound reported to be doped into the metallic state is the two-leg ladder compound $La_{1-x}Sr_xCuO_{2.5}$, and no superconductivity was observed.[14]

As in $(VO)_2P_2O_7$, an estimate of Δ in the two-leg ladder

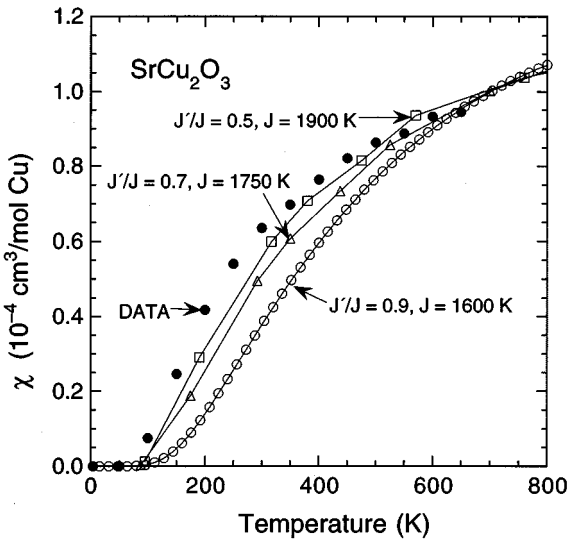

FIG. 1. Representative magnetic spin susceptibility χ vs temperature data for $SrCu_2O_3$ (Ref. 9, filled circles). Also shown as open symbols and connecting lines are fits to these data by the calculations of Barnes and Riera for the isolated spin-$\frac{1}{2}$ two-leg ladder (Ref. 6) for several ratios of the rung to leg exchange constants J'/J and for the fitted values of J.

compound $SrCu_2O_3$ (Ref. 9) has been made from $\chi(T)$ measurements. A few representative $\chi(T)$ data for $SrCu_2O_3$ (Ref. 9) are plotted in Fig. 1. To obtain a value for Δ, Azuma *et al.*[9] fitted their $\chi(T)$ data by the prediction of Troyer *et al.*[25] for isolated spin-$\frac{1}{2}$ ladders,

$$\chi(T)=\frac{A}{\sqrt{T}}e^{-\Delta/k_BT}, \tag{2}$$

where k_B is Boltzmann's constant. A very good fit was obtained (not shown in Fig. 1) with $\Delta/k_B = 420$ K, and with the A value in Table I. Assuming that $J\approx J'$, as usually done up to now when considering spin-$\frac{1}{2}$ ladder compounds and found as cited above for $(VO)_2P_2O_7$, one has $\Delta\approx J/2$.[3,4,19,20,25] In this case one obtains $J/k_B\approx 840$ K for $SrCu_2O_3$.[9] Surprisingly, this value is considerably less than the range $J/k_B = 1700$–3000 K estimated[26–31] for the linear-CuO_3-chain compound Sr_2CuO_3 and also much less than values of ~ 1500 K found[32] in the layered cuprate superconductor parent compounds.

Herein, we first analyze in Sec. II the fits to and the magnitudes of $\chi(T)$ as reflected in the value of A, assuming that the interladder exchange coupling is negligible. We show that the experimental A value for $SrCu_2O_3$ (Ref. 9) in Table I is not consistent with the assumption that $J\approx J'$ for isolated ladders, but rather indicates that $J'/J\lesssim 0.5$. From fits of the $\chi(T)$ data by the calculations of Barnes and Riera,[6] we confirm this estimate. The assumption of isolated ladders also leads to the conclusion that the AF coupling along the ladder legs in this compound is very strong ($J\gtrsim 2000$ K). The influence of interladder coupling on $\chi(T)$ is discussed in Secs. III and IV. In Sec. III, a general method is presented for estimating exchange constants in unfrustrated AF quantum Heisenberg spin-$\frac{1}{2}$ systems from the maximum spin susceptibility $\chi^{\max}$ value when the effective *magnetic* coordination number $z_{\rm eff}$ is unknown or unclear, such as can happen, for example, in spin-ladder compounds if the strength of the interladder coupling is not negligible. From this treatment and the experimental data, bounds are placed on the inter- and intraladder coupling constants in $SrCu_2O_3$. In Sec. IV, using a mean-field approach and the results in Sec. III, an expression for $\chi(T)$ incorporating the influence of interladder coupling on $\chi(T)$ is derived and utilized to fit the data for $SrCu_2O_3$. The inferred values of J'/J and J are similar to the above values obtained by assuming negligible interladder coupling. Concluding remarks are given in Sec. V.

II. MAGNETIC SUSCEPTIBILITY OF ISOLATED SPIN-$\frac{1}{2}$ TWO-LEG LADDERS

The general form for the ''magnon'' dispersion relation of an isolated spin-$\frac{1}{2}$ two-leg ladder near the minimum at wave vector $k=\pi/a$ is given by

$$\varepsilon(ka)=\Delta+\gamma(\delta ka)^2, \tag{3}$$

TABLE I. Parameters describing the lowest triplet excited state dispersion relation for two-leg spin-$\frac{1}{2}$ ladders (see text). A number in parentheses in the lower table is the estimated standard deviation in the last digit of the preceding quantity. The units of A are 10^{-3} $cm^3\,K^{1/2}$/mol Cu. Equation numbers refer to the text.

Compound	$\frac{\Delta}{k_B}$ (K)	A	Ref.	$\frac{\gamma}{k_B}$ (10^4 K)	γ/Δ	Eq.
$SrCu_2O_3$	420	4	9	1.4	33	(5)

$J'J$	$\varepsilon(0)/J$	Δ/J	Theory c_o	$\varepsilon(0)/\Delta$	γ/Δ	Eq., Ref.
2.0	3.148(1)	1.280(3)	0.59(3)	2.46(1)	0.90(2)	(10), 4
1.0	1.89(1)	0.51(3)	0.88(2)	3.7(2)	5.3(3)	(10), 4
					4.9	(8)
$\varepsilon^{(1)}(k)$		0.496			6.39	25
$\varepsilon^{(2)}(k)$		0.517			5.07	25
0.5	1.09(4)	0.30(16)	1.02(3)	3.7(19)	16.(5)	(10), 4
					24.	(8)

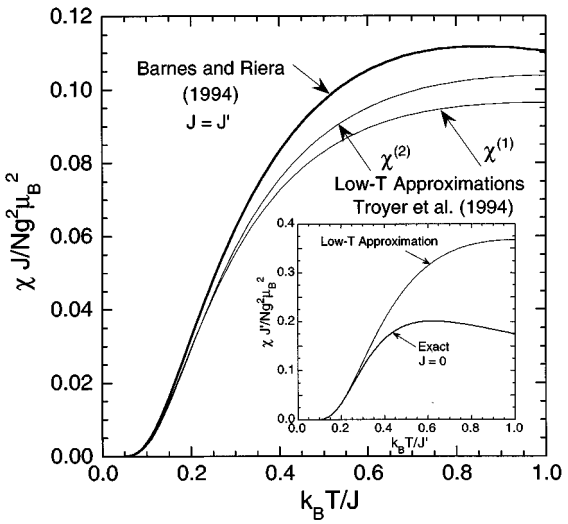

FIG. 2. Magnetic spin susceptibility χ vs temperature T computed for the spin-$\frac{1}{2}$ two-leg ladder with $J=J'$ (Ref. 6). Also shown are extrapolations to high T of two low-T approximations of Troyer *et al.* (Ref. 25). In the inset are shown the computed $\chi(T)$ for a spin-$\frac{1}{2}$ dimer in Eq. (6) and the extrapolated low-T approximation.

where $\delta k \equiv k-\pi/a$ in the extended zone scheme and the parameter γ is the curvature at the band minimum as defined by Eq. (3). Using this form, Troyer *et al.*[25] obtained an expression for $\chi(T)$ in the low temperature limit, $k_BT \ll \Delta$ and $k_BT \ll$ magnon bandwidth, given by

$$\chi(T)=\frac{Ng^2\mu_B^2}{2k_B\sqrt{\pi(\gamma/k_B)T}}\exp(-\Delta/k_BT), \tag{4}$$

where N is the number of spins 1/2, g is the Landé factor, and μ_B is the Bohr magneton. For $N=1$ mole (Avogadro's number), one obtains Eq. (2), with

$$A=\frac{g^2(0.1058\ \mathrm{cm^3\ K^{1/2}/mol})}{\sqrt{\gamma/k_B}}. \tag{5}$$

Several issues are important regarding fits of $\chi(T)$ data by Eq. (2) for two-leg ladder compounds. The first is that Eq. (2) is derived for the low-T limit, whereas for $SrCu_2O_3$,[9] for example, the data and fit extended to a temperature of $1.55\Delta/k_B$. It is thus not clear how the experimental parameters A and Δ are related to the calculated ones in Eqs. (2) and (5) when data are fitted over an extended T range. At present, calculations are not available to address this issue for arbitrary J'/J. However, as shown in the inset of Fig. 2, if $J=0$ ($\Delta=J'$), corresponding to isolated rung dimers, the low-T approximation $\chi_L(T)=[Ng^2\mu_B^2/k_BT]e^{-\Delta/k_BT}$ seriously diverges from the exact result

$$\chi(T)=\frac{Ng^2\mu_B^2}{k_BT}\,\frac{e^{-\Delta/k_BT}}{1+3e^{-\Delta/k_BT}} \tag{6}$$

for $T \gtrsim 0.3J'/k_B$. In contrast, as shown in Fig. 2, the two low-T approximations of Troyer *et al.*[25] for $J=J'$ [for dispersion relations $\varepsilon^{(1)}(ka)$ and $\varepsilon^{(2)}(ka)$ with the Δ and γ parameters in Table I] are found using Eq. (4) to differ by $\lesssim 10\%$ from the accurate calculations[6,25] for at least $k_BT/J \lesssim 1$, corresponding[3,4,20,25] to $k_BT/\Delta \lesssim 2$.

Second, as will be seen below, for comparison with theory it is important to be able to determine the parameter γ, in addition to Δ, from the experimental $\chi(T)$ data. From Eq. (4), this in turn requires that the g factor be known or at least quantitatively estimated. To our knowledge, for Cu^{+2} in square-planar oxygen coordination, with or without apical oxygens, the (anisotropic) g factor is always ≥ 2,[26] as is the case in, e.g., La_2CuO_4 as determined from $\chi(T)$ data[33] and $YBa_2Cu_3O_{6+\delta}$ from NMR data.[34] This observation originates from the negative sign of the spin-orbit coupling parameter for Cu^{+2} and from the crystalline electric field effects on the d-orbital energies. Thus, the g value is not an arbitrarily adjustable parameter. An average g value of about 2.1 is consistent with the maximum observed ordered moment ($\sim 0.7\mu_B$) in the Néel state of the AF insulator layered cuprate parent compounds.[32] We therefore question recent analyses of $\chi(T)$ data yielding $g \ll 2$, in the absence of justification for these g values, for Sr_2CuO_3 ($g=1.6$) (Ref. 27) and $SrCu_2O_3$ ($g=1.4$–1.55).[35] Since the g factor comes into $\chi(T)$ calculations as the square, such g values reduce the calculated $\chi(T)$ (and J, see below) by roughly a factor of 2 from those calculated using $g \gtrsim 2$. In the remainder of this paper, g will be taken to be the fixed value 2.1.[26]

The third issue to be addressed is the magnitude of $\chi(T)$, which is reflected in the value of A in Eq. (2). From Eq. (5), the A value for $SrCu_2O_3$ yields the γ value in Table I. The experimental Δ value in Table I then gives $\gamma/\Delta \approx 33$. This ratio is about a factor of 5 larger than predicted for the case $J=J'$ for the $\varepsilon^{(1)}(k)$ and $\varepsilon^{(2)}(k)$ dispersion relations of Troyer *et al.*,[25] as shown in Table I. With decreasing J'/J, Δ/J decreases.[3,4] Also, as J'/J decreases, the curvature γ at the magnon band minimum increases.[4,6,19] From these considerations, one expects the ratio γ/Δ to increase with decreasing J'/J. The fact that the observed γ/Δ ratio for $SrCu_2O_3$ is much larger than those predicted for $J'/J=1$ thus suggests that J'/J is significantly less than 1 in this compound.

An estimate is now made of γ/Δ versus J'/J for $J'/J \leq 1$. The dispersion relation for a single isolated chain is[36] $\varepsilon^0(ka)=(\pi J/2)|\sin(ka)|$. In the presence of a spin gap Δ, the modified dispersion relation near $ka=\pi$ is assumed to be $\varepsilon(ka) \approx \{\Delta^2+[\varepsilon^0(ka)]^2\}^{1/2}$. Expanding $\varepsilon^0(ka)$ for small deviations δk of k from π/a then gives $\gamma/\Delta \approx (1/8)(\pi J/\Delta)^2$. The Δ/J versus J'/J results of Barnes *et al.*[4] for the bulk limit and for $J'/J \leq 1$ are well fitted by

$$\Delta/J=0.4(J'/J)+0.1(J'/J)^2, \tag{7}$$

as shown in Fig. 3. Combining the last two expressions gives

$$\frac{\gamma}{\Delta} \approx \frac{7.71(J/J')^2}{[1+0.25(J'/J)]^2}. \tag{8}$$

Using Eq. (8), the γ/Δ ratio is predicted to be 4.9 for $J'/J=1$, which is close to the estimates for $J'/J=1$ by Troyer *et al.*[25] in Table I. For $J'/J=0.5$, Eq. (8) gives $\gamma/\Delta \approx 24$.

The $\varepsilon(ka)$ for the 2×12 spin-$\frac{1}{2}$ two-leg ladder has been calculated using Lanczos techniques by Barnes and Riera[6]

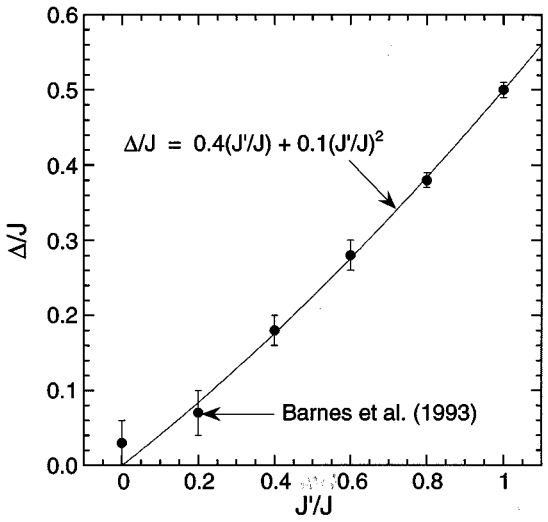

FIG. 3. Computed spin gap Δ vs J'/J for the spin-$\frac{1}{2}$ two-leg ladder, in the region $J'/J \leq 1$ (Ref. 4). The solid curve is a polynomial fit of order 2, forced to pass through (0,0), to the data.

for $J'/J = 2$, 1 and 0.5. For $J'/J = 2$ and 1, their values $\Delta/J \equiv \varepsilon(\pi)/J = 1.28$ and 0.52, respectively, are in good agreement with respective bulk limit calculations.[3,4,19,20,25] For $J'/J = 0.5$, their value of 0.29 is about 30% larger than the bulk value in Fig. 3,[4] as expected.[3,4] Estimates of γ/Δ versus J'/J can be obtained from the $\varepsilon(ka)$ calculations. Barnes and Riera found that their data could be fitted well by

$$\varepsilon(ka) = [\varepsilon^2(0)\cos^2(ka/2) + \varepsilon^2(\pi)\sin^2(ka/2) + c_o^2(\pi J/2)^2 \sin^2(ka)]^{1/2}, \quad (9)$$

where $\Delta \equiv \varepsilon(\pi)$. For $J'/J \to 0$, one must have $\varepsilon(0) \to 0$, $\varepsilon(\pi) \to 0$, $c_o \to 1$. Our fitting parameters of the $\varepsilon(ka)$ calculations are listed in Table I. For ka close to the band minimum at π, Eq. (9) yields

$$\frac{\gamma}{\Delta} = \frac{1}{8}\left[\left(\frac{\varepsilon(0)}{\Delta}\right)^2 - 1 + \left(\frac{\pi c_o J}{\Delta}\right)^2\right]. \quad (10)$$

The values of γ/Δ obtained from Eq. (10) and from the $\varepsilon(0)$, Δ and c_o parameters in Table I are listed in Table I. The γ/Δ ratio is seen to increase strongly with decreasing J'/J. For $J'/J = 1$, the γ/Δ ratio is comparable with those of Troyer *et al.* and our estimate using Eq. (8) in Table I. The γ/Δ value for $J'/J = 0.5$ for the 2×12 ladder is somewhat smaller than our estimate from Eq. (8) for the bulk limit.

The above estimates of γ/Δ and comparison with the experimental value in Table I suggest that $J'/J \lesssim 0.5$ in $SrCu_2O_3$. Equation (8) and the observed γ/Δ ratio in Table I suggest that $0.4 < J'/J < 0.5$ in this compound.

Numerical calculations of $\chi(T)$ exist for J'/J values down to 0.5.[6,25,35] The theoretical $\chi(T)$ predictions for $J'/J < 1$ by Barnes and Riera[6] were therefore scaled onto the experimental data[9] for $SrCu_2O_3$ in Fig. 1. For each value of J'/J, J was varied until agreement with at least the highest T experimental data was obtained, as shown in Fig. 1. As anticipated above, values of $J'/J \geq 0.9$ are clearly ruled out, with the lowest available value $J'/J = 0.5$ providing the best (but still not optimum) fit. The evolution of the fits with decreasing J'/J indicates that J'/J is somewhat less than 0.5, and that $J \gtrsim 2000$ K.

The J'/J ratio derived for $SrCu_2O_3$ from consideration of the γ/Δ ratios in Table I ($\lesssim 0.5$) is consistent with the independent theoretical fits to the $\chi(T)$ data in Fig. 1. In addition, using $J'/J = 0.5$ and $J = 2000$ K from above, Eq. (7) yields $\Delta/k_B = 450$ K, similar to the experimentally inferred value of 420 K in Table I. We note that the calculations of Gopalan, Rice, and Sigrist[19] for the ''trellis'' spin lattice in $SrCu_2O_3$ indicated for $J'/J = 1$ that Δ for each individual ladder is unaffected by the frustrated intralayer interladder interactions. However, the strength of interlayer interladder interactions was not investigated, but may be significant and important to the interpretation of the $\chi(T)$ data,[37] an issue which we now address.

III. ANALYSIS OF THE MAXIMUM SPIN SUSCEPTIBILITY FOR SPIN-$\frac{1}{2}$ HEISENBERG ANTIFERROMAGNETS

In this section, we consider the information that can be gained about the AF exchange coupling constants between the spins of a material from the measured maximum value $\chi^{\max}$ of $\chi(T)$, assuming nearest-neighbor Heisenberg exchange interactions only. The spin Hamiltonian for a general system is written

$$\mathcal{H} = \sum_{\langle i,j \rangle} J_{ij} \boldsymbol{S}_i \cdot \boldsymbol{S}_j, \quad (11)$$

where the sum is over unique nearest-neighbor pairs and $J_{ij} > 0$ denotes an AF interaction. Here the discussion is limited to bipartite (two-sublattice) magnetic lattices in which an ordered collinear AF (Néel) state can be imagined where all the nearest neighbors of a member of one sublattice belong to the second sublattice (other cases will be briefly discussed at the end of this section). If all J_{ij} are the same ($\equiv J$) and each spin has the same number z of nearest neighbors, molecular field theory (MFT) predicts[38] that

$$\frac{\chi^{\max} J z}{N g^2 \mu_B^2} = \frac{1}{2}, \quad (12)$$

independent of spin S. One can generalize Eq. (12) to systems in which the magnetic environment of each spin is the same but where the J_{ij} are not equal. Since in Eq. (12) $Jz = \Sigma_j J_{ij}$, where j runs over all nearest neighbors of a given spin i, we define an effective magnetic coordination number $z_{\rm eff}$ by

$$z_{\rm eff} = \frac{1}{J^{\max}} \sum_j J_{ij}, \quad (13)$$

where $J^{\max} = \max(J_{ij})$. Then, Eq. (12) becomes

$$\frac{\chi^{\max} J^{\max} z_{\rm eff}}{N g^2 \mu_B^2} = \frac{1}{2}. \quad (14)$$

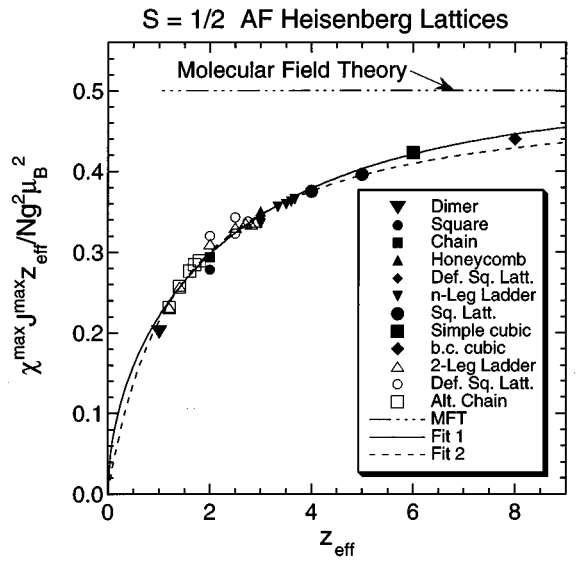

FIG. 4. Computed maximum spin susceptibility $\chi^{\max} \equiv \max[\chi(T)]$ times the effective magnetic coordination number $z_{\rm eff}$ vs $z_{\rm eff}$ from the literature for various spin-$\frac{1}{2}$ Heisenberg antiferromagnets (see text). Fits 1 (solid curve) and 2 (dashed curve) are fits to the data by Eqs. (15) and (17) in the text, respectively.

Of course, MFT neglects strong fluctuation (dynamic short-range AF order) effects which increase with decreasing S and with decreasing z or $z_{\rm eff}$. Accurate calculations and measurements are expected, and found, to give a $\chi^{\max}$ smaller than predicted in Eqs. (12) and (14). One might expect a systematic variation of $\chi^{\max} J^{\max} z_{\rm eff}/Ng^2\mu_B^2$ with $z_{\rm eff}$ for a given S, with the deviation from the MFT prediction of 1/2 decreasing with increasing $z_{\rm eff}$.

To test these ideas, calculations of $\chi^{\max}$ from the literature for various spin-$\frac{1}{2}$ systems are presented in Fig. 4 as a plot of $\chi^{\max} J^{\max} z_{\rm eff}/Ng^2\mu_B^2$ versus $z_{\rm eff}$. A systematic variation is indeed observed versus $z_{\rm eff}$, with the deviation from the MFT prediction decreasing with increasing $z_{\rm eff}$, as expected. Included in Fig. 4 are $\chi^{\max}$ data for lattices with isotropic J (i.e., $J_{ij}=J$, filled symbols) including the dimer ($z=1$) from Eq. (6), the linear chain ($z=2$),[28] the planar honeycomb lattice ($z=3$),[39] the ordered defect square lattice ($z=3$) of CaV_4O_9,[40] the two-leg ladder ($z=3$),[6,25] the square lattice ($z=4$),[32,39,41] the two-layer square lattice ($z=5$), the simple-cubic lattice ($z=6$), and the body-centered-cubic lattice ($z=8$).[39] For the isotropic nonfrustrated n-leg ladders with $n=3$, 4, 5, and 6,[42] $z_{\rm eff}$ is defined to be the average coordination number of a spin in a ladder: $z_{\rm eff}\equiv 4-(2/n)$. In addition, lattices with anisotropic J_{ij} (open symbols) are included in Fig. 4: the alternating-exchange linear chain in which two different J_{ij} alternate along the chain ($z_{\rm eff}=1.2$–1.8),[6] the anisotropic two-leg ladder ($z_{\rm eff}=1.2$–2.9),[6,25] and the anisotropic ordered defect square lattice of CaV_4O_9 ($z_{\rm eff}=2$–2.75).[40] The solid curve in Fig. 4 (''Fit 1'') is a fit of all of the data by the empirical expression

$$\frac{\chi^{\max} J^{\max} z_{\rm eff}}{Ng^2\mu_B^2} = \frac{1}{2}\tanh(0.477 z_{\rm eff}^{0.521}). \qquad (15)$$

Equation (15) becomes identical with the MFT result (14) in the limit of large $z_{\rm eff}$. Remarkably, it is seen from Fig. 4 that $\chi^{\max}$ depends primarily on $z_{\rm eff}$, and is not sensitive (to within $\sim 10\%$) to the space dimensionality of the spin lattice or to the magnitude of any spin-gap relative to $J^{\max}$.

To illustrate the utility of the theoretical calculations in Fig. 4 [and/or of the fit to those data in Eq. (15)], consider the $\chi(T)$ data for $SrCu_2O_3$ in Fig. 1. It appears that $\chi^{\max}$ is given to within a few percent by $\chi^{\max}=1.0\times10^{-4}$ cm^3/mol Cu (this is confirmed below in Sec. IV). Then Fig. 4 and/or Eq. (15) yields the following quantitative insights. First suppose that interladder spin exchange coupling is negligible. If the exchange coupling within the ladder is isotropic, then $z=3$ ($=z_{\rm eff}$) and $J/k_B=1900$ K; this estimate of J/k_B is more than a factor of 2 larger than the estimate of $\approx$840 K cited in the Introduction assuming isotropic ladders. If the intrachain interaction $J=0$, then $z=1$ and the rung exchange constant $J'/k_B=$ 3300 K. On the other hand, if $J'=0$, corresponding to isolated chains with $z=2$, then the intrachain $J/k_B=2460$ K. If $J'/J=1/2$, then $J^{\max}=J$, $z_{\rm eff}=2.5$, $J/k_B=2140$ K and $J'/k_B=1070$ K; these values are consistent with those found above in Sec. II for $SrCu_2O_3$.

Now suppose that interladder spin exchange coupling is not negligible. For example, suppose that $z_{\rm eff}=4$ (implying a strong interladder spin exchange coupling) in $SrCu_2O_3$, which would correspond to an isotropic square lattice, or, e.g., to an isotropic two-leg ladder with intraladder exchange constant J and where each Cu spin in a ladder is coupled to a Cu spin in each of two adjacent ladders with exchange constant $J/2$. Then one obtains $J=1560$ K, again still much larger than the estimate of 840 K in the Introduction but similar to the known exchange coupling constants in other similar cuprates. To proceed further and determine which of the above or other possibilities actually applies requires fits to the $\chi(T)$ data by specific models. This was done assuming no interladder exchange coupling in Sec. II. Fits assuming non-negligible interladder coupling will be presented in Sec. IV.

Finally, the influence of possible magnetic frustration on the phenomenology in Fig. 4 is briefly discussed. All of the $\chi^{\max}$ data in that figure are for nonfrustrated bipartite AF spin lattices. An example of a frustrated lattice is the (close-packed) planar triangular lattice with $z=6$. For an isotropic $S=1/2$ Heisenberg antiferromagnet on this lattice, Elstner *et al.* find that $\chi^{\max} zJ/Ng^2\mu_B^2=0.536$,[43] significantly larger than the value in Fig. 4 for $z=6$, indicating that the geometric frustration suppresses the development of dynamic short-range AF order. Other frustrated lattices include isolated clusters of N spins ($N>2$) in which each spin interacts with every other spin in the cluster with the same exchange constant J, where $z=N-1$. The energy of such a cluster is given exactly in terms of the total spin S of the cluster by $E_S=(J/2)S(S+1)$, apart from an additive constant, where for N even and constituent spins-$\frac{1}{2}$, one has $S=0, 1, \ldots, N/2$. The energy level degeneracies and $\chi(T)$ to arbitrary accuracy are easily evaluated.[44,45] We find $\chi^{\max} zJ/Ng^2\mu_B^2=0.4024$ for $z=3$, 0.5065 for $z=5$, 0.5715 for $z=7, \ldots$. These values are again larger than those for the respective $z_{\rm eff}$ values in Fig. 4. Therefore, for given values of $\chi^{\max}$ and $z_{\rm eff}$, the value of $J^{\max}$ obtained from Fig. 4 or Eq. (15) is evidently a lower limit.

IV. TEMPERATURE DEPENDENCE OF THE SPIN SUSCEPTIBILITY OF QUASI-LOW-DIMENSIONAL HEISENBERG SPIN SYSTEMS

A. General considerations

We now consider the relevance of the theoretical data in Fig. 4 in determining the influence of intersystem spin couplings on the *temperature-dependent* spin susceptibility $\chi(T)$ of the total coupled system. In particular, suppose one has an isolated spin system with $z_{\rm eff}\equiv z_0$ and an accurately known spin susceptibility $\chi_0(T)$. In the absence of an explicit accurate calculation of $\chi(T)$ for coupled systems, one must resort to some sort of mean-field theory to calculate it. Such theories often give the general form $\chi(T)=\chi_0(T)/[1+\lambda\chi_0(T)]$, where λ is independent of T. Since this expression is presumed to hold at each T, it must hold in particular for the temperature $T^{\rm max}$ at which χ is a maximum, which allows us to make contact with the theoretical results in Fig. 4. This expression is consistent with Fig. 4 if, as will be shown in the next paragraph, it is written as

$$\chi(T)=\frac{\chi_0(T)}{1+2(z_{\rm eff}-z_0)[\chi_0(T)J^{\rm max}/Ng^2\mu_B^2]}. \qquad (16)$$

The strength of intersystem exchange coupling is contained in the parameter $z^*\equiv z_{\rm eff}-z_0$. Equation (16) will be used below to fit the experimental $\chi(T)$ data for $SrCu_2O_3$, where in this case $\chi_0(T)$ is the susceptibility of an isolated spin-$\frac{1}{2}$ two-leg ladder.

After multiplying both sides of Eq. (16) by $J^{\rm max}z_{\rm eff}/Ng^2\mu_B^2$ and setting $T=T^{\rm max}$, one obtains

$$\frac{\chi^{\rm max}J^{\rm max}z_{\rm eff}}{Ng^2\mu_B^2}=\frac{\chi_0^{\rm max}J^{\rm max}z_{\rm eff}/Ng^2\mu_B^2}{1+2(z_{\rm eff}-z_0)(\chi_0^{\rm max}J^{\rm max}/Ng^2\mu_B^2)}. \qquad (17)$$

The dashed curve in Fig. 4 (''Fit 2'') is a fit of all the theoretical data points in Fig. 4 by Eq. (17) for $z_0=1$; the fit is seen to be accurate to about 10% for all of the data points and much better than this for most of them. Essentially the same fit was obtained for $1\leqslant z_0\leqslant 5$. In the limit of large $z_{\rm eff}$, Eq. (17) yields the MFT result, Eq. (14); this is in fact the criterion by which the factor of 2 (rather than, e.g., a more precise value for a given fitting range of $z_{\rm eff}$) was inserted in the denominators on the right-hand-sides of Eqs. (16) and (17).

B. $SrCu_2O_3$

In this section, fits by Eq. (16) to the experimental $\chi(T)$ data in Fig. 1 for $SrCu_2O_3$ will be presented. For $\chi_0(T)$, the calculations of Barnes and Riera[6] for $J'/J=1$, 0.7 and 0.5 are utilized. Before giving the results, we point out that Eq. (16) is expected to be accurate when fitting $\chi(T)$ data for which $\chi\lesssim\chi^{\rm max}$ since Eq. (16) was derived for this regime. However, the accuracy of Eq. (16) is unclear for the range $\chi\ll\chi^{\rm max}$. For example, the spin gap decreases when nonfrustrated interladder coupling exists,[17] which is expected to cause the lowest-temperature behavior of $\chi(T)$ to be significantly different than predicted by the mean-field expression (16). Therefore, Eq. (16) was fitted to the higher-temperature ($\gtrsim$300 K) experimental data; agreement of the extrapolated fit with the lower-temperature data, when it occurs, is tentatively considered to be fortuitous.

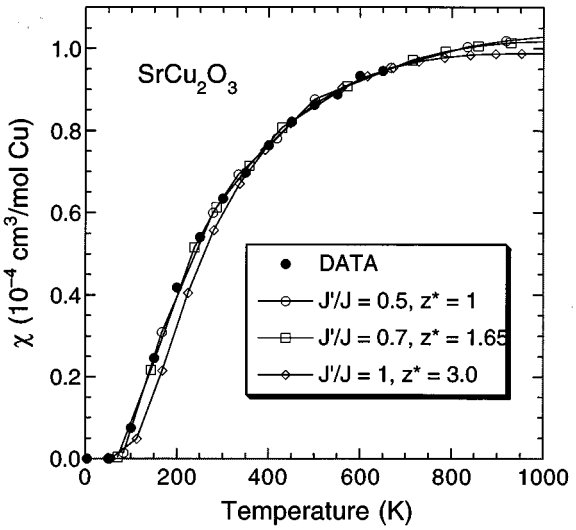

FIG. 5. Measured spin susceptibility χ vs temperature for $SrCu_2O_3$ (Ref. 9), as in Fig. 1. Theoretical fits to the data assuming the existence of interladder coupling, by Eq. (16) in the text and using the calculated spin susceptibilities of isolated two-leg spin-$\frac{1}{2}$ ladders of Barnes and Riera for $J'/J=0.5$, 0.7, and 1, (Ref. 6) are shown by open symbols and connecting lines.

The fits by Eq. (16) to the experimental $\chi(T)$ data[9] in Fig. 1 for $SrCu_2O_3$ are shown in Fig. 5. An essentially perfect fit to the high-T data was obtained for each of the three ratios of J'/J. For $J'/J=1$ ($z_0=3$), the best fit gave $z^*\equiv z_{\rm eff}-z_0=3.0(3)$ and $J/k_B=1120(30)$ K. The value of z^* (and consequently J'/J and J) is not acceptable, since it corresponds to each Cu spin in the sample having the same unfrustrated coupling to each of $z_{\rm eff}=6$ nearest neighbors, equivalent to a simple-cubic spin lattice in three space dimensions. Such a system would not exhibit the strong short-range AF ordering over such a large temperature range as observed, but rather would exhibit long-range AF order at $T_N\sim J/k_B$.[39] For $J'/J=0.7$ ($z_0=2.7$), the values $z^*=1.65$ and $J/k_B=1430$ K were obtained. Finally, for $J'/J=0.5$ ($z_0=2.5$), the parameters are $z^*=1.0(2)$ and $J/k_B=1900(200)$ K. Unfortunately, $\chi_0(T)$ calculations are not available for $J'/J<0.5$, so our fits could not be extended into this parameter regime. Since $SrCu_2O_3$ exhibits a spin-liquid ground state as discussed in the Introduction, the z^* value for $J'/J=0.7$ seems too large to be realistic. Thus, we tentatively come to similar conclusions reached in Sec. II, that $J'/J\sim 0.5$ and $J/k_B\sim 2000$ K in $SrCu_2O_3$.

V. CONCLUDING REMARKS

The analysis in Sec. II of $\chi(T)$ for $SrCu_2O_3$,[9] in terms of isolated ladders, suggests that $J'/J\lesssim 0.5$ in this two-leg spin-ladder compound. The AF coupling along the legs of the two-leg ladders is found to be very strong, $J\gtrsim 2000$ K, similar to previous estimates for the linear chain compound Sr_2CuO_3.[26–30] The inferred J'/J and J values are consistent with the observed spin-gap and with the general discussion

of $\chi^{\max}$ in Sec. III; the results of this section and Sec. IV A may be generally useful in analyzing $\chi(T)$ data for other quasi-low-dimensional spin systems. From Sec. IV, the above parameters appear to be supported even after inclusion of interladder coupling in the mean-field-type fits to the experimental $\chi(T)$ data.

The inferred suppression of J' with respect to J in $SrCu_2O_3$ is unexpected and needs to be further tested and investigated by microscopic calculations and additional experiments. If confirmed, this anisotropy may have a significant influence on the predicted physical properties of the metallic doped spin-ladder compounds. For example, this suppression may reduce the possibility of superconductivity due to an electronic mechanism (although this reduction may be ameliorated by the large value of J),[1,3,24] and may help to explain why no superconductivity was observed in the metallic spin-$\frac{1}{2}$ two-leg ladder $La_{1-x}Sr_xCuO_{2.5}$ system.[14] Further accurate numerical calculations of $\chi(T)$ for isolated ladders are needed for $J'/J<0.5$ to extend the available range of predictions for comparison with the data. Most importantly, the accuracy of the parameters obtained from our mean-field-type fits in Sec. IV to the experimental data needs to be determined by accurate numerical calculations of the influence of interladder exchange coupling on $\chi(T)$ for $J'/J \lesssim 1$.

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