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MAGNETIC SUSCEPTIBILITY OF Sr_2CuO_3 AND $SrCu_2O_3$

D.C. JOHNSTON

Ames Laboratory-USDOE and Department of Physics and Astronomy
Iowa State University
Ames, Iowa 50011, USA

The observed magnetic susceptibility $\chi^{obs}(T)$ data of Ami et al. for the linear-chain cuprate Sr_2CuO_3 are re-analyzed in terms of recent theory of Eggert et al. for the spin susceptibility $\chi(T)$ of the spin $S = 1/2$ linear-chain Heisenberg antiferromagnet, which yielded the Cu–Cu exchange constant $J/k_B = 2150$ K. Values for J', the exchange constant in the rungs, and J, that in the legs, were estimated for the $S = 1/2$ two-leg ladder compound $SrCu_2O_3$, using the $\chi(T)$ data of Azuma et al. The analyses suggest that $J'/J \approx 0.5$, contrary to the expectation that $J'/J \approx 1$, and that J/k_B is very large (≈ 2000 K), similar to that in Sr_2CuO_3.

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1. Introduction

Spin configurations formed by coupling n spin chains side-by-side in a plane are termed n-leg spin ladders, where the coupling between spins is antiferromagnetic (AF). Such spin ladders have received increasing attention over the last several years (for an excellent review, see Ref. [1]). Experimental research on spin ladders was stimulated by theoretical predictions that the (nonfrustrated) spin $S = 1/2$ two-leg ladder should have a nonmagnetic quantum "spin-liquid" ground state, with a spin gap to the lowest magnetic triplet excited states, in contrast to the isolated chain which has no spin gap. This prediction was verified for the $S = 1/2$ two-leg ladder compounds $(VO)_2P_2O_7$ (Refs. [2–4]) and $SrCu_2O_3$ [5]. For wider ladders, the 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. The latter prediction was verified for the three-leg ladder compound $Sr_2Cu_3O_5$, which showed no spin gap [5] and exhibited disordered static AF ordering below ≈ 50 K [6]. 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. For AF Heisenberg exchange interactions between nearest-neighbor (nn) spins, the only case discussed here, the spin Hamiltonian of the n-leg spin ladder is

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

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

Herein, we first present an analysis of the observed magnetic susceptibility vs. temperature $\chi^{\rm obs}(T)$ data [7] for the prototypical $S = 1/2$ linear-chain antiferromagnet Sr_2CuO_3 in terms of the recent theoretical predictions of Eggert et al. for the spin susceptibility $\chi(T)$ of the $S = 1/2$ linear chain nn Heisenberg antiferromagnet [8]. We find that the exchange energy J between Cu^{2+} spins is large, $J/k_{\rm B} \approx 2150$ K, where $k_{\rm B}$ is Boltzmann's constant. A general method is then presented for estimating exchange constants in nonfrustrated $S = 1/2$ AF quantum Heisenberg systems from the maximum spin susceptibility $\chi^{\rm max}$ value. From this treatment and the experimentally extracted $\chi(T)$ data [5], bounds are placed on the inter- and intraladder exchange constants in $SrCu_2O_3$. We then review the analysis [9] of the $\chi(T)$ data [5] for $SrCu_2O_3$ in terms of the calculations of Barnes and Riera [3], which assume isolated ladders. The assumption of isolated ladders leads to the conclusion that $J' \lesssim J/2$ and that J is very large ($J/k_{\rm B} \gtrsim 2000$ K), as in Sr_2CuO_3. The influence of interladder coupling on $\chi(T)$ is then discussed [9]. An expression for $\chi(T)$ incorporating the influence of interladder coupling 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.

2. Magnetic susceptibility of Sr_2CuO_3

The powder $\chi^{\rm obs}(T)$ data [7] for Sr_2CuO_3 are shown in Fig. 1. These data were previously analyzed in terms of the Bonner–Fisher calculation [10] for $\chi(T)$ of the $S = 1/2$ linear chain Heisenberg antiferromagnet, which yielded the large value $J/k_{\rm B} = 2600^{+200}_{-400}$ K [7]. A more accurate calculation of $\chi(T)$, which is significantly different than that of Bonner and Fisher at $T \lesssim 0.2J/k_{\rm B}$, has since become available [8]. We therefore refitted the $\chi^{\rm obs}$ data by Eggert the expression

$$\chi^{\rm obs}(T) = \chi_0 + \frac{C_i}{T + \theta_i} + \chi(T), \tag{2}$$

where χ_0 is a temperature-independent orbital contribution, the second term is a Curie–Weiss impurity/defect contribution, and $\chi(T)$ is the calculation of Eggert et al. [8]. The best fit of Eq. (2) to the data is shown as the heavy solid curve in Fig. 1, where $\chi_0 = -7.5 \times 10^{-5}$ cm^3/mol, $C_i = 4.0 \times 10^{-4}$ cm^{3}K/mol, $\theta_i = 4.5$ K, and $J/k_{\rm B} = 2150^{+150}_{-100}$ K assuming [7] a Landé factor $g = 2.1$ for the bulk spins. The value of $J/k_{\rm B}$ is significantly larger than observed in the layered cuprate superconductor parent compounds (≈ 1500 K), but is similar to the value (2200±200) K obtained from a recent identical analysis of $\chi^{\rm obs}(T)$ data for a single crystal [11], where however $g = 2$ was assumed (H. Eisaki, private communication). Eggert fitted the data in Fig. 1 using the same theory [8] as used here and the same

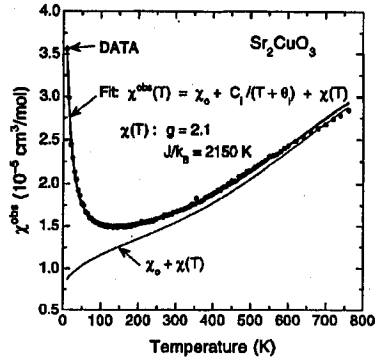

Fig. 1. Observed magnetic susceptibility χ^{obs} vs. temperature data for the linear chain compound Sr_2CuO_3 [7]. The solid curve through the data is a theoretical fit (see text).

expression (2), and obtained $J/k_B = 1700^{+150}_{-100}$ K [12], where (S. Eggert, private communication) $g = 1.6$ for Cu^{2+} was used; this g-value seems unrealistically low, since to our knowledge Cu^{2+} always shows $g \geq 2$ in oxides. A value $J/k_B \approx 3000$ K was obtained from optical measurements on Sr_2CuO_3 [13]. Theoretical calculations indicate that J/k_B can be no larger than about 2300 K in this compound [14].

3. $\chi(T)$ of $S = 1/2$ Heisenberg antiferromagnets

Every specific antiferromagnet has a characteristic χ value associated with it, namely the value $\chi^{max} \equiv \max[\chi(T)] \equiv \chi(T^{max})$. Here, we consider the information which can be gained about the AF exchange coupling constants between the spins of a material from the measured χ^{max} value, assuming nn Heisenberg exchange interactions only. The treatment is somewhat different from the one we gave previously [9]. To motivate the form of the expression to be used to analyze published calculations of $\chi(T)$ (and χ^{max}) for various spin lattices, and also to provide preliminaries needed for Sec. 4, we first review the Weiss molecular field theory (MFT) for $\chi(T)$.

Suppose we have a magnetic system consisting of coupled subsystems, where the spin susceptibility $\chi_0(T)$ of an isolated subsystem is accurately known. In MFT, the thermal average $\langle \cdots \rangle$ of the magnetic moment μ_i of a given spin i in the coupled system is $\langle \mu_i \rangle = \chi_0' H_{eff}$, where H_{eff} is the effective magnetic induction seen by the spin i, and χ_0' is the susceptibility *per spin* of the isolated subsystem. One writes H_{eff} as $H_{eff} = H + H_{ex}$, where H is the applied magnetic field and H_{ex} is the (average) exchange field seen by μ_i due to exchange coupling with its neighboring magnetic moments μ_j. In MFT, one assumes that

$$H_{ex} = -\lambda \langle \mu_j \rangle, \tag{3}$$

where $\lambda > 0$ is the AF molecular field coupling constant. Combining the last three expressions gives $\langle \mu_i \rangle = \chi_0'(H - \lambda \langle \mu_j \rangle)$. We assume that all spins are magnetically equivalent, therefore in the paramagnetic state one has $\langle \mu_j \rangle = \langle \mu_i \rangle$. Then, solving for $\langle \mu_i \rangle$ and using the definitions $\chi' \equiv \langle \mu_i \rangle / H$, $\chi = N\chi'$ and $\chi_0 = N\chi_0'$ where N

is the number of spins in the system, one obtains

$$\frac{1}{\chi(T)} = \frac{1}{\chi_0(T)} + \frac{\lambda}{N}. \tag{4}$$

We now express λ in terms of the exchange coupling constants J_{ij}^* between a spin in a given subsystem $\boldsymbol{S}_i$ and spins $\boldsymbol{S}_j$ in adjacent subsystems. The spin Hamiltonian for the coupled system is $\mathcal{H} = \mathcal{H}_0 + \mathcal{H}^*$, where the first term is for an isolated subsystem and the second is the interaction Hamiltonian between subsystems. The latter Hamiltonian is written $\mathcal{H}^* = \sum_{\langle i,j \rangle} J_{ij}^* \boldsymbol{S}_i \cdot \boldsymbol{S}_j$, where the sum is over nn bonds and $J_{ij}^* > 0$ denotes an AF interaction. Using the definition $\boldsymbol{\mu} = -g\mu_B \boldsymbol{S}$ where μ_B is the Bohr magneton, one has $\mathcal{H}^* = \sum_i \boldsymbol{\mu}_i \cdot \sum_j (J_{ij}^*/g^2\mu_B^2)\boldsymbol{\mu}_j$. Writing the average exchange energy E_{ex}^* between $\boldsymbol{\mu}_i$ and its intersubsystem neighbors as $E_{ex}^* = \langle \mu_i \rangle \langle \mu_j \rangle \sum_j (J_{ij}^*/g^2\mu_B^2) \equiv -\langle \mu_i \rangle H_{ex}$, one has

$$H_{ex} = -\langle \mu_j \rangle \sum_j \frac{J_{ij}^*}{g^2\mu_B^2}. \tag{5}$$

Comparing Eq. (5) with (3) yields

$$\lambda = \frac{\sum_j J_{ij}^*}{g^2\mu_B^2}. \tag{6}$$

Inserting Eq. (6) into (4) then gives

$$\frac{1}{\chi(T)} = \frac{1}{\chi_0(T)} + \frac{\sum_j J_{ij}^*}{Ng^2\mu_B^2}. \tag{7}$$

If a subsystem consists of a single spin, then $\chi_0(T)$ is the Curie-law susceptibility of isolated spins, $\chi_0(T) = C/T$, where the Curie constant C is given by $C = Ng^2\mu_B^2 S(S+1)/3k_B$. Substituting this χ_0 into Eq. (7) then yields the Curie–Weiss law, $\chi(T) = C/(T+\theta)$, where $\theta = S(S+1)\sum_j J_{ij}^*/3k_B$. If all of the J_{ij}^* are the same, $\equiv J$, and the number of nearest neighbors is defined as z, then

$$\sum_j J_{ij}^* = zJ \tag{8}$$

and one obtains the familiar form $\theta = S(S+1)zJ/3k_B$.

By analogy with Eq. (8), in general when the exchange coupling constants J_{ij} of a given spin i to its neighbors j are not all the same, we define an *effective magnetic coordination number*, z_{eff}, by the relation $z_{eff}J^{max} \equiv \sum_j J_{ij}$, or

$$z_{eff} = \frac{1}{J^{max}} \sum_j J_{ij}, \tag{9}$$

where $J^{max} \equiv \max(J_{ij})$, i.e., J^{max} is the largest exchange coupling constant in the system.

Now for a system composed of interacting subsystems as discussed above, one can write

$$\sum_j J_{ij} = \sum_j J_{ij}^0 + \sum_j J_{ij}^*, \tag{10}$$

where the first sum on the right hand side is over nearest neighbors of spin i within a subsystem and the second sum is over nearest neighbors of spin i between subsystems. We define the effective magnetic coordination number within a subsystem as $z_0 \equiv \sum_j J_{ij}^0/J^{\max}$ and between subsystems as $z^* \equiv \sum_j J_{ij}^*/J^{\max}$. Then Eq. (9) becomes

$$z_{\text{eff}} = z_0 + z^*. \tag{11}$$

To simplify notation, we also define

$$\chi^* \equiv \frac{\chi J^{\max}}{Ng^2\mu_B^2}. \tag{12}$$

Upon multiplying both sides of Eq. (7) by $Ng^2\mu_B^2/J^{\max}$, and using Eqs. (9) and (11), one obtains the simple forms

$$\frac{1}{\chi^*(T)} = \frac{1}{\chi_0^*(T)} + \alpha z^* = \frac{1}{\chi_0^*(T)} + \alpha(z_{\text{eff}} - z_0), \tag{13}$$

where in this case $\alpha = 1$.

Setting $T = T^{\max}$ in Eq. (13) yields

$$\frac{1}{\chi^{*\max}} = \frac{1}{\chi_0^{*\max}} + \alpha(z_{\text{eff}} - z_0), \tag{14}$$

where again $\alpha = 1$. Equations (13) and (14) were derived using MFT, and are therefore only expected to be valid in the weak-coupling limit

$$z^* = z_{\text{eff}} - z_0 \ll 1. \tag{15}$$

In this limit, from Eq. (14) a plot of $1/\chi^{*\max}$ vs. z_{eff} should give a straight line with a slope of 1. Examination of published numerical calculations of $\chi^*(T)$ for a number of $S = 1/2$ systems consisting of weakly-coupled subsystems shows that this is indeed correct in each case as long as $z^* = z_{\text{eff}} - z_0 \lesssim 0.1$. This analysis will be published separately. Here, as will be seen below, we are interested in the more strongly coupled regime in which the condition (15) is not satisfied. In this case, we expect Eq. (14) to hold on average, but where the constant α is to be empirically determined.

Shown in Fig. 2 is a plot of $1/\chi^{*\max}$ vs. z_{eff} for a number of geometrically nonfrustrated 0D, 1D, 2D and 3D $S = 1/2$ lattices for which $1 \leq z_{\text{eff}} \leq 8$. Included in Fig. 2 are $\chi^{*\max}$ data for lattices with isotropic J (i.e., $J_{ij} = J$), including the dimer ($z = 1$), the square cluster ($z = 2$), the linear chain ($z = 2$) [8], the planar honeycomb lattice ($z = 3$) [15], the ordered-vacancy square lattice ($z = 3$) of CaV_4O_9 [16], the two-leg ladder ($z = 3$) [3], the square lattice ($z = 4$), the two-layer square lattice ($z = 5$), the simple-cubic lattice ($z = 6$) and the body-centered-cubic lattice ($z = 8$) [15]. For the n-leg ladders with $n = 3$, 4, 5 and 6 [17], z_{eff} is defined to be the average coordination number of a spin in a ladder: $z_{\text{eff}} \equiv 4 - 2/n$. In addition, lattices with anisotropic J_{ij} are included in Fig. 2: the alternating-exchange linear chain in which two different J_{ij} alternate along the chain ($z_{\text{eff}} = 1.2$–1.8) [3], the anisotropic two-leg ladder ($z_{\text{eff}} = 1.2$–2.9) [3, 18], and the anisotropic ordered-vacancy square lattice of CaV_4O_9 ($z_{\text{eff}} = 2$–2.75) [16]. On the global scale of the figure, all of the data can be fitted reasonably well by a straight line, confirming Eq. (14):

$$\frac{1}{\chi^{*\max}} = 2.83 + 1.93\, z_{\text{eff}}. \tag{16}$$

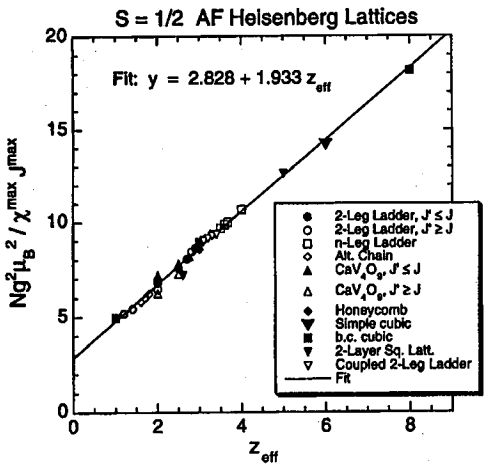

Fig. 2. Inverse of the maximum spin susceptibility $\chi^{\max}$ from calculations vs. the effective magnetic coordination number $z_{\rm eff}$ for various $S = 1/2$ Heisenberg antiferromagnets (see text). The straight line is a linear fit to all of the data.

Referring to Eq. (14), the slope is $\alpha = 1.93$. The above MFT predicts that $T^{\max} = \theta$, yielding $\alpha = 2$, which is close to the value derived from Fig. 2. Inserting the definition of χ^* in Eq. (12) into Eq. (16), and using $N =$ Avogadro's number and $g = 2.1$, typical of the average g value of Cu^{2+} spins-1/2 in oxides, one obtains

$$\chi^{\max} = \frac{1.65\ \text{cm}^3\,\text{K/mol Cu}}{(2.83 + 1.93 z_{\rm eff})(J^{\max}/k_{\rm B})}. \tag{17}$$

Thus, from the measured $\chi^{\max}$ value, one can obtain an estimate of the possible range of exchange coupling constants in a compound.

4. Analysis of $\chi(T)$ of $SrCu_2O_3$

4.1. Determination of exchange constants from the $\chi^{\max}$ value

To illustrate the use of Eq. (17), we apply it to the case of the $S = 1/2$ two-leg ladder compound $SrCu_2O_3$. Representative $\chi(T)$ data [5] for this compound are shown in Fig. 3. It appears that $\chi^{\max}$ is given to within a few percent by $\chi^{\max} = 1.0 \times 10^{-4}$ cm^3/mol Cu (confirmed below). Then Eq. (17) yields the following quantitative insights. First suppose that interladder spin exchange coupling is negligible. If the exchange coupling within the ladder is spatially isotropic, then $z = 3$ ($= z_{\rm eff}$), leading to $J/k_{\rm B} = 2000$ K. If the intrachain interaction $J = 0$, corresponding to isolated dimers, then $z = 1$ and the rung exchange constant $J'/k_{\rm B} = 3600$ K. On the other hand, if $J' = 0$, corresponding to isolated chains with $z = 2$, then the intrachain $J/k_{\rm B} = 2500$ K. If $J'/J = 1/2$, then $J^{\max} = J$ and $z_{\rm eff} = 2.5$, so that Eq. (17) yields $J/k_{\rm B} = 2200$ K and $J'/k_{\rm B} = 1100$ K.

Now suppose that interladder spin exchange coupling is not negligible. For example, suppose that $z_{\rm eff} = 5$ (implying a strong interladder spin exchange coupling) in $SrCu_2O_3$, which would correspond, e.g., to an isotropic two-leg ladder

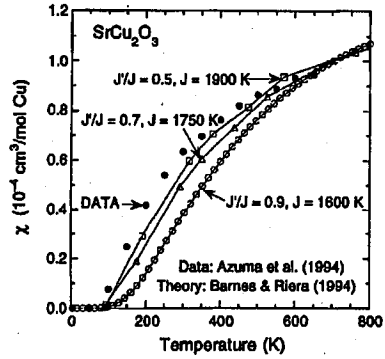

Fig. 3. Representative magnetic spin susceptibility χ vs. temperature data for $SrCu_2O_3$ (filled circles) [5]. The open symbols and connecting lines are fits to these data by calculations for the isolated $S = 1/2$ two-leg ladder [3], for several ratios of the rung to leg exchange constants J'/J and for the fitted values of J [9].

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 the same exchange constant J. Then one obtains $J/k_B = 1400$ K.

Thus, we conclude that a lower limit on J^{max}/k_B in $SrCu_2O_3$ is 1400 K. To proceed further and determine which of the above or other possibilities actually applies requires fits to the $\chi(T)$ data by specific models, which we now attempt.

4.2. Analysis assuming isolated ladders

Numerical calculations of $\chi(T)$ for isolated $S = 1/2$ two-leg Heisenberg ladders exist for J'/J values down to 0.5 [3]. The theoretical $\chi(T)$ predictions for $J'/J < 1$ by Barnes and Riera [3] were therefore scaled onto the $\chi(T)$ data [5] for $SrCu_2O_3$ in Fig. 3. 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. 3. Values of $J'/J \geq 0.9$ are clearly ruled out, with the lowest value $J'/J = 0.5$ providing the best (but still not optimum) fit. The evolution of the fits with decreasing J'/J indicates that $J'/J < 0.5$ and $J/k_B \gtrsim 2000$ K.

4.3. Analysis assuming coupled ladders

Here, we utilize Eqs. (12) and (13), and set α to the MFT value of 2. For $\chi_0(T)$, the calculations of Barnes and Riera [3] for $J'/J = 1$, 0.7 and 0.5 are utilized. The fits by Eq. (13) to the experimental $\chi(T)$ data [5] in Fig. 3 for $SrCu_2O_3$ are shown in Fig. 4. An essentially perfect fit to the high-T ($\gtrsim 300$ K) data was obtained for each of the three ratios of J'/J. For $J'/J = 1$ ($z_0 = 3$), the best fit gave $z^* \equiv z_{eff} - z_0 = 3.0(3)$ 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 $z_{eff} = 6$ nearest neighbors, equivalent to a simple cubic spin lattice. Such a system would not exhibit the strong short-range AF ordering over such a large temperature range

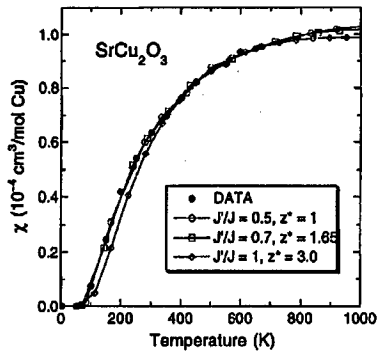

Fig. 4. χ vs. temperature for $SrCu_2O_3$ [5], as in Fig. 3. Theoretical fits to the data assuming interladder coupling are shown by open symbols and connecting lines [9].

below T^{max} as observed, but rather would exhibit long-range AF order at $T_N \sim J/k_B$ [15]. 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. Since $SrCu_2O_3$ exhibits a spin-liquid ground state as discussed in Sec. 1, the z^* value for $J'/J = 0.7$ seems too large to be realistic. Thus, we tentatively come to similar conclusions reached above, that $J'/J \approx 0.5$ and $J/k_B \approx 2000$ K in $SrCu_2O_3$.

5. Concluding remarks

The analysis in Sec. 4 of $\chi(T)$ for $SrCu_2O_3$ assuming isolated ladders suggests that $J'/J \lesssim 0.5$ in this two-leg spin ladder compound. The AF coupling along the legs of the ladders is found to be very strong, $J/k_B \gtrsim 2000$ K, similar to our and others' estimates for the $S = 1/2$ linear chain compound Sr_2CuO_3, $J/k_B \approx 2200$ K. The inferred J'/J and J values are consistent with the general discussion of χ^{max} in Sec. 3; the results of this section may be generally useful in analyzing $\chi(T)$ data for other spin systems. From Sec. 4, the above parameters for $SrCu_2O_3$ appear to be supported even after inclusion of interladder coupling in the mean-field type fits to the $\chi(T)$ data. Further theoretical work is needed to improve the accuracy of the derived parameters.

Acknowledgments

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References

[1] E. Dagotto, T.M. Rice, *Science* **271**, 618 (1996).

[2] D.C. Johnston, J.W. Johnson, D.P. Goshorn, A.J. Jacobson, *Phys. Rev. B* **35**, 219 (1987).

[3] T. Barnes, J. Riera, *Phys. Rev. B* **50**, 6817 (1994).

[4] R.S. Eccleston, T. Barnes, J. Brody, J.W. Johnson, *Phys. Rev. Lett.* **73**, 2626 (1994).

[5] M. Azuma, Z. Hiroi, M. Takano, K. Ishida, Y. Kitaoka, *Phys. Rev. Lett.* **73**, 3463 (1994).

[6] K. Kojima, A. Keren, G.M. Luke, B. Nachumi, W.D. Wu, Y.J. Uemura, M. Azuma, M. Takano, *Phys. Rev. Lett.* **74**, 2812 (1995).

[7] T. Ami, M.K. Crawford, R.L. Harlow, Z.R. Wang, D.C. Johnson, Q. Huang, R.W. Erwin, *Phys. Rev. B* **51**, 5994 (1995).

[8] S. Eggert, I. Affleck, M. Takahashi, *Phys. Rev. Lett.* **73**, 332 (1994).

[9] D.C. Johnston, *Phys. Rev. B*, to be published.

[10] J.C. Bonner, M.E. Fisher, *Phys. Rev.* **135**, A640 (1964).

[11] N. Motoyama, H. Eisaki, S. Uchida, *Phys. Rev. Lett.* **76**, 3212 (1996).

[12] S. Eggert, *Phys. Rev. B* **53**, 5116 (1996).

[13] H. Suzuura, H. Yasuhara, A. Furusaki, N. Nagaosa, Y. Tokura, *Phys. Rev. Lett.* **76**, 2579 (1996).

[14] S.-L. Drechsler, J. Málek, S. Zališ, K. Rościszewski, *Phys. Rev. B* **53**, 11328 (1996).

[15] For a review see R. Navarro, in: *Magnetic Properties of Layered Transition Metal Compounds*, Ed. L.J. de Jongh, Kluwer, Dordrecht 1990, p. 105.

[16] M. Troyer, H. Kontani, K. Ueda, *Phys. Rev. Lett.* **76**, 3822 (1996).

[17] B. Frischmuth, B. Ammon, M. Troyer, *Phys. Rev. B* **54**, R3714 (1996).

[18] M. Troyer, H. Tsunetsugu, D. Würtz, *Phys. Rev. B* **50**, 13515 (1994).