Accurate determination of the exchange constant in Sr_2CuO_3 from recent theoretical results

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Data from susceptibility measurements [Ami *et al.*, Phys. Rev. B **51**, 5994 (1995)] on Sr_2CuO_3 are compared with recent theoretical predictions [Eggert *et al.*, Phys. Rev. Lett. **73**, 332 (1994)] for the magnetic susceptibility of the antiferromagnetic spin-1/2 Heisenberg chain. The experimental data fully confirms the theoretical predictions and in turn we establish that Sr_2CuO_3 behaves almost perfectly like a one-dimensional antiferromagnet with an exchange coupling of $J=1700^{+150}_{-100}$ K.

The Hamiltonian for the antiferromagnetic spin-1/2 Heisenberg chain

$$H=J\sum_i \vec{S}_i\cdot\vec{S}_{i+1} \tag{1}$$

has been a popular and very well studied model in theoretical physics for a very long time. It was not until recently, however, that the bulk susceptibility per site

$$\chi(T)\equiv\frac{g^2\mu_B^2}{k_BT}\sum_i \langle S_0^z S_i^z\rangle_T \tag{2}$$

was calculated for the full temperature range by combining analytical arguments from field theory and numerical results from the algebraic Bethe ansatz equations.[2] That work has to be viewed in relation to the pioneering results of Bonner and Fisher,[3] who calculated the susceptibility of finite chains numerically more than 30 years ago. The results of Bonner and Fisher correctly predicted a broad maximum in the susceptibility $\chi(T)$ which was very useful for determining the characteristics of quasi-one-dimensional materials with moderate values of J.[4,5] Attempts to extrapolate the Bonner and Fisher data to lower temperatures,[6] however, turned out to yield incorrect results.

The most surprising result of Ref. 2 is the prediction of a divergent slope of $\chi(T)$ at $T=0$ together with an inflection point at $T\approx0.087J$. At low temperatures the deviation of the extrapolated Bonner and Fisher curve from this exotic behavior is quite significant as shown in Fig. 1. The value at zero temperature is $\chi(0)=1/J\pi^2$ (we set the gyromagnetic ratio times the Bohr magneton $g\mu_B$ as well as the Boltzmann constant k_B to unity, so that the susceptibility is measured in units of $1/J$ and the temperature is measured in units of J).

We will now attempt to present experimental evidence for the surprising behavior of the susceptibility at low temperatures. The one-dimensional characteristics of all experimental systems break down at some finite temperature (because of a spin-Peierls transition or three-dimensional ordering), but nonetheless we expect that the best quasi-one-dimensional materials should show an inflection point at low temperatures and a significant deviation from the extrapolated Bonner and Fisher curve.

The material Sr_2CuO_3 is believed to have the best one-dimensional characteristics of an antiferromagnet reported so far.[1] In particular, it is one of the few materials which will be able to exhibit the difference between the two curves in Fig. 1. (In fact, it is probably the only known material to have a low enough transition temperature besides $CuCl_2\cdot 2NC_5H_5$, for which a clear deviation at low temperatures from the Bonner and Fisher curve was first noticed in the early 70's.[5]) Sr_2CuO_3 is very much of current interest since it is directly related to high-temperature superconductors and $Sr_2CuO_{3.1}$ was reported to exhibit high-temperature superconductivity ($T_c\approx70$ K) under high presure.[7] The exchange constant J in Eq. (1) is expected to be roughly the same as the Cu-Cu superexchange interaction in the layered cuprates since the Cu-Cu distances are comparable.

A number of susceptibility measurements have been performed on this material[8,9] and rather good results are available from recent measurements on carefully prepared, high-quality samples of Sr_2CuO_3 by Ami *et al.*[1] Their data was analyzed under the assumption that an extrapolated Bonner and Fisher curve yielded good results also for lower temperatures. They reported a general agreement with the Bonner and Fisher curve and an exchange constant of $J=2600^{+200}_{-400}$ K.

We took the identical experimental data and performed a

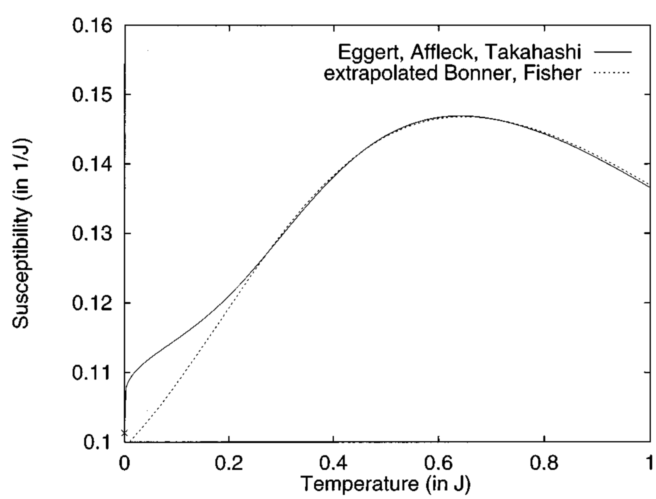

FIG. 1. The susceptibility of the spin-1/2 chain according to recent calculations (Ref. 2) compared to the extrapolated Bonner and Fisher curve (from Ref. 6).

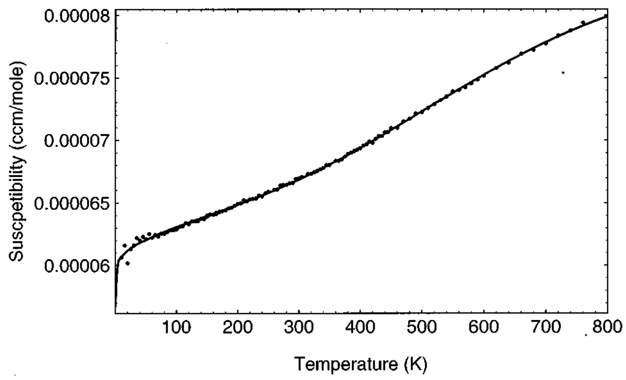

FIG. 2. The fit of the magnetic susceptibility according to the results of Ref. 2 with $J=1700$ K. The background susceptibility and the Curie-Weiss term are subtracted.

fit according to the newly available data from the numerical Bethe ansatz calculations of Ref. 2 but using otherwise identical assumptions. Namely, after subtracting the core diamagnetism (from Ref. 10), we fitted the total susceptibility χ^{tot} assuming a constant term from Van Vleck paramagnetism χ^{VV}, a Curie-Weiss term per impurity $\chi^{\text{CW}}(T) = g^2\mu_B^2 S(S+1)/3k_B(T-\Theta)$, and the spin chain part $\chi(T)$ from Ref. 2:

$$\chi^{\text{tot}}(T) = \chi(T) + \rho\chi^{\text{CW}}(T) + \chi^{\text{VV}}, \tag{3}$$

where ρ is the impurity density (assuming nearly isolated finite length spin chains with an odd number of spins).

The result of our fit is shown in Fig. 2 which yielded a dramatically different estimate for the exchange constant

$$J = 1700^{+150}_{-100} \text{ K} \tag{4}$$

compared to $J=2600$ K in Ref. 1 (taking into account their different definition of J by a factor of 2). Other parameters in our fit are $\theta \approx -4.49$ K, a Van Vleck susceptibility of $\chi^{\text{VV}} \approx 2.55 \times 10^{-5}$ ccm/mole and an impurity density of $\rho \approx 0.16\%$.

Our greatly different estimate of J, however, is based on a much better fit of the experimental data. The deviation of the experimental data from the least-squares fit is plotted in Fig. 3 for two different cases:

(A) our fit according to Ref. 2 ($J=1700$ K),

(B) the fit according to the extrapolated Bonner and Fisher curve ($J=2614$ K), [fit (B) was taken directly from Ref. 1 which in turn was based on Refs. 3 and 6].

We can see that the fit (B) to the extrapolated Bonner and Fisher curve contains a systematic deviation which cannot be explained by experimental error. Our new fit (A), however, is fully within the statistical fluctuations of the experimental data. Both fits have random deviations at very low temperatures that are larger than the scale of the graph (3), which may be due to larger experimental error in that region. In any case we cannot expect that a Curie-Weiss term is fully adequate to describe impurity effects in that temperature region. However, this has little effect on the fit over the full temperature range or on the estimate of J.

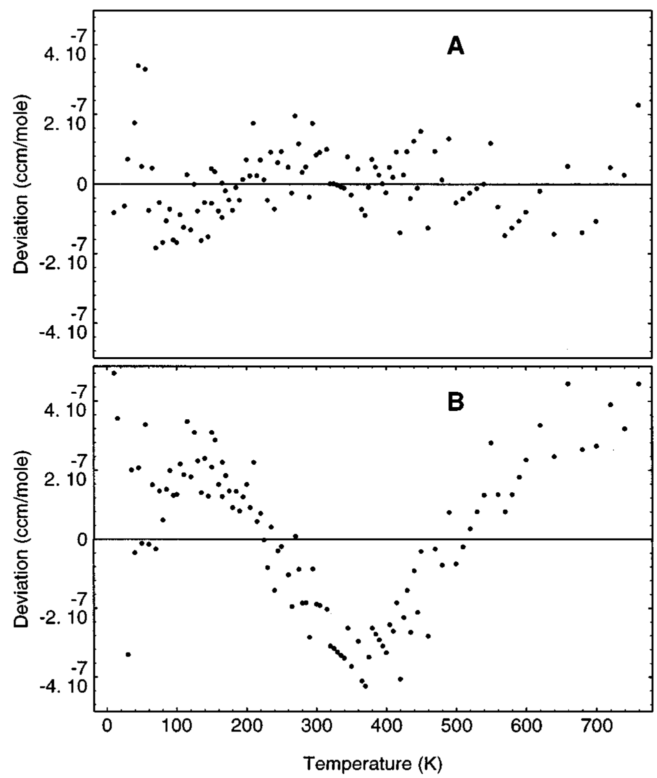

FIG. 3. The deviation of the experimental data from the least-squares fit using the results of Eggert *et al.* (Ref. 2) (A) and using the extrapolated Bonner and Fisher result (Ref. 6) (B).

We take the extremely good fit as strong evidence that Ref. 2 predicted the susceptibility of the one-dimensional Heisenberg model correctly. Moreover, the quality of the fit establishes that Sr_2CuO_3 is very well described by the model in Eq. (1) over a large temperature range. There has been no report of a spin-Peierls transition in this material and the three-dimensional ordering temperature was reported to be $T_N \approx 5$ K from μSR experiments.[9] This makes Sr_2CuO_3 the antiferromagnet with the best quasi-one-dimensional characteristics reported so far $J/T_N \approx 300$.

In conclusion we have presented strong experimental confirmation for the predicted exotic temperature dependence of the Heisenberg chain susceptibility at low temperatures.[2] The material Sr_2CuO_3 has been established as a highly one-dimensional antiferromagnet with a much improved estimate of the exchange constant $J = 1700^{+150}_{-100}$ K which compares rather well with the values of the exchange interaction in the layered cuprates [$\approx 1480 \pm 80$ K (Ref. 111)]. An independent experimental check of the exchange constant J in Sr_2CuO_3 would certainly be desirable.

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