Comment on "Spin Correlations in the Paramagnetic Phase and Ring Exchange in La_2CuO_4"

In a recent Letter, Toader *et al.* [1] claimed to provide definitive experimental evidence for ring-exchange terms in the Hamiltonian of La_2CuO_4 by comparing the experimental antiferromagnetic static spin structure factor $S(\mathbf{Q})$ with high-temperature series expansion. Ring-exchange terms arise at intermediate coupling in the effective low-energy theory for the Hubbard model. The parameters deduced from Ref. [1] agree with those found earlier [2], and for the Hubbard model they correspond to $\kappa \equiv t/U = 1/7.5$, with $t = 0.29$ eV.

Figure 1 compares the high-temperature series expansions of $S(\mathbf{Q})$ in Fig. 3 of Ref. [1] represented by the two dashed lines, along with three of the experimental points in the upper right-hand corner, with our quantum Monte Carlo (QMC) data for $U = 7.5t$ and $U = 10t$ (the dotted lines are a guide to the eye). We used the determinantal QMC method with discretization step $\Delta\tau = 1/10$. When a size independent value is reached, we give as lower and upper error bars statistical fluctuations on, respectively, the smallest and largest result. For the lowest temperatures, up to $L \times L = 12 \times 12$, the size dependence is important and the upper error bar is obtained by a $1/L$ extrapolation for the two largest system sizes. Since $J/(2T) = \frac{1}{2}(J/t)(t/T) \equiv (\kappa/2)\beta$, the horizontal scale depends only on the ratio κ, for temperatures in units of t. We add as a dash-dotted line the value of $S(\mathbf{Q})$ obtained from high-temperature series expansion of the Heisenberg model without ring exchange [3]. The Heisenberg and QMC results, computed with $\hbar = 1$, are scaled by the same factor to compare with Ref. [1] where the origin of the vertical log scale is arbitrary. As expected, the larger the value of U, the better the agreement between the Heisenberg and the Hubbard models. Note, however, that both values of U are much closer to the Heisenberg result than to those of Ref. [1], but that $U = 7.5t$ should correspond to the exchange parameters (including ring exchange) used in that reference.

In Ref. [1] it was argued that agreement with experiment was obtained with ring exchange because the high-temperature series results dovetail better with the experiment than the results obtained without ring exchange. We have here a counterexample since the $U = 7.5t$ or $U = 10t$ Hubbard model results can both smoothly join the experimental data, if we use the appropriate vertical scale. As an added observation, we show in Fig. 1 that with $J/t = 2\tilde{J}_2^{(1)}/t = 4\kappa - 64\kappa^3$, as suggested in Ref. [1], all QMC results (symbols without error bars) fall close to the Heisenberg curve which joins smoothly the experiment.

The experimental results on $S(\mathbf{Q})$ can be described by the nonlinear sigma model (universal regime), and hence

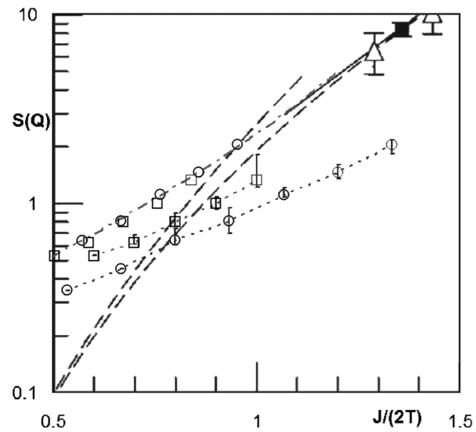

FIG. 1. High-temperature series expansion from Ref. [1]: lower dashed line is fifth order; higher dashed line is fourth order. Experimental results in upper right-hand corner. Heisenberg model [3], dash-dotted line. QMC results: $U = 7.5t$, circles; $U = 10t$, squares. When no error bars are indicated, there is additional scaling, as discussed in the text.

they are insensitive to microscopic details, as noted in Ref. [1]. Our results suggest that, unless this $S(\mathbf{Q})$ can be measured at higher temperature, a smoothness argument cannot lead to an accurate value of U/t (and hence of the ring-exchange contribution), especially if we allow for further neighbor hoppings. Moreover, even with measurements of $S(\mathbf{Q}, \omega)$ [2], one also needs detailed information on the band structure to get reliable values of U and consequent ring-exchange terms [4].

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