Comment on “Spin Correlations in the Paramagnetic Phase and Ring Exchange in La$_2$CuO$_4$”

In a recent Letter, Toader et al. [1] claimed to provide definitive experimental evidence for ring-exchange terms in the Hamiltonian of La$_2$CuO$_4$ by comparing the experimental antiferromagnetic static spin structure factor $S(Q)$ 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 $\kappa = t/U = 1/7.5$, with $t = 0.29$ eV.

Figure 1 compares the high-temperature series expansions of $S(Q)$ 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 $J/(2T) = (J/t)(t/T) \equiv (\kappa/2)\beta$, the horizontal scale depends only on the ratio $\kappa$, for temperatures in units of $t$. We add as a dash-dotted line the value of $S(Q)$ obtained from high-temperature series expansion of the Heisenberg model without ring exchange [3]. The Heisenberg and QMC results, computed with $h = 1$, 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 $J/t = 2J^{(1)}_2/t = 4\kappa - 64\kappa^3$, 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 $S(Q)$ can be described by the nonlinear sigma model (universal regime), and hence they are insensitive to microscopic details, as noted in Ref. [1]. Our results suggest that, unless this $S(Q)$ 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 $S(Q, \omega)$ [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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