Effects of intrabilayer coupling on the magnetic properties of

YBa$_2$Cu$_3$O$_6$

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Abstract

A two-layer Heisenberg antiferromagnet is studied as a model of the bilayer cuprate YBa$_2$Cu$_3$O$_6$. Quantum Monte Carlo results are presented for the temperature dependence of the spin correlation length, the static structure factor, the magnetic susceptibility, and the $^{63}$Cu NMR spin-echo decay rate $1/T^2_G$. As expected, when the ratio $J_2/J_1$ of the intrabilayer and in-plane coupling strengths is small, increasing $J_2$ pushes the system deeper inside the renormalized classical regime. Even for $J_2/J_1$ as small as 0.1 the correlations are considerably enhanced at temperatures as high as $T/J_1 \approx 0.4 - 0.5$. This has a significant effect on $1/T^2_G$, and it is suggested that measurements of this quantity at high temperatures can reveal the strength of the intrabilayer coupling in YBa$_2$Cu$_3$O$_6$. 
One of the unresolved issues regarding the high-$T_c$ cuprate superconductors is the role of the coupling between CuO$_2$ planes within the same “block” in bi- and tri-layer compounds. Suggestions$^1$ that this coupling might be responsible for the spin-gap behavior observed$^3$ in the bilayer cuprate YBa$_2$Cu$_3$O$_{6.6}$ has spurred recent theoretical work on models of two coupled CuO$_2$ planes.$^4$–$^6$ Experimentally, the strength of the intrabilayer coupling in YBa$_2$Cu$_3$O$_{6+x}$ has not yet been accurately determined. Neutron scattering experiments performed in the insulating regime have not detected the high-energy branch of the spin-fluctuation spectrum up to energies of 60meV.$^7$ This negative result analyzed in the framework of a linear spin-wave theory suggests a lower bound of $J_2 = 8$ meV.$^7$ In the recently synthesized high-$T_c$ superconductor YBa$_4$Cu$_7$O$_{15}$ the two planes constituting a bilayer have slightly different chemical environments,$^8$ enabling various cross-relaxation experiments.$^9$ Analyzing such experiments by Stern et al.$^{10}$ Millis and Monien estimated 5 meV $< J_2 < 20$ meV.$^6$ On the other hand, quantum chemical calculations indicate a value of $J_2$ in YBa$_2$Cu$_3$O$_{6+x}$ as high as $\approx50$ meV.$^{11}$ In this situation, it is important to consider new possible experiments that could accurately determine the magnitude of $J_2$.

Here we explore possibilities of extracting the strength of the intrabilayer coupling from experiments in the antiferromagnetic insulating regime, e.g. for YBa$_2$Cu$_3$O$_6$. We report results of quantum Monte Carlo (QMC) simulations of a two-layer Heisenberg model defined by the hamiltonian

$$\hat{H} = J_1 \sum_{a=1,2} \sum_{\langle i,j \rangle} \vec{S}_{ai} \cdot \vec{S}_{aj} + J_2 \sum_i \vec{S}_{1i} \cdot \vec{S}_{2i}, \tag{1}$$

where $\vec{S}_{ai}$ is a spin-$1/2$ operator at site $i$ of layer $a$, and $\langle i,j \rangle$ denotes a pair of nearest-neighbor sites on a square lattice. This model should be a reasonable starting point for describing the magnetic properties of YBa$_2$Cu$_3$O$_6$ at temperatures higher than the Neel temperature, which in clean samples is as high as 500K. Since the in-plane coupling $J_1 \approx 1200K,$$^7$ temperatures of interest are $0.4 \lesssim T/J_1 \lesssim 0.5$. We present results in this regime for the spin-spin correlation length, the static structure factor, the susceptibility, and the $^{63}$Cu NMR spin-echo decay rate $1/T_{2G}$, and discuss the possibilities to experimentally detect the
influence of a weak $J_2$.

The finite-temperature physics of two-dimensional quantum antiferromagnets was explained in the framework of a mapping onto a nonlinear $\sigma$-model some time ago by Chakravarty, Halperin and Nelson.\cite{12} If the ground state is ordered the correlation length $\xi$ in the low-temperature renormalized classical (RC) regime diverges as $e^{2\pi \rho_s/T}$, where $\rho_s$ is the spin-stiffness constant. At temperatures $T \gg \rho_s$, $T$ is the only relevant energy scale and the behavior is quantum critical (QC), with $\xi \sim T^{-1}$.\cite{12,13} If $\rho_s$ is large, the cross over boundary between the RC and QC regimes of the $\sigma$-model may be at a temperature where a $\sigma$-model description of the antiferromagnet is no longer valid, and instead there is a direct cross-over from the RC to a local moment (LM) regime where the spins are essentially decoupled.

For a single Heisenberg plane ($J_2/J_1 = 0$), the behavior at the temperatures of interest here is influenced by all the above regimes. For $0.4 \lesssim T/J_1 \lesssim 0.6$ there is a cross-over from RC to QC behavior, and at higher temperatures LM effects influence the behavior in the narrow QC regime.\cite{14,15} For the two-layer model described by (1), the ground state order increases with $J_2$ for small $J_2/J_1$, and has a maximum around $J_2/J_1 \approx 0.8$ before decreasing due to the tendency to singlet formation across the planes for larger $J_2$.\cite{16} A quantum phase transition to a disordered ground state occurs at $(J_2/J_1)_c \approx 2.5$.\cite{17,18} The finite-temperature properties of near-critical systems have been studied numerically in detail, enabling a direct verification of the applicability of $1/N$ calculations for the nonlinear $\sigma$-model in the QC regime.\cite{18,19} Here we are concerned with values of $J_2/J_1$ more reasonable for modeling high-$T_c$ bilayer cuprates, and choose $J_2/J_1 = 0.1, 0.2,$ and $0.5$. Owing to the enhanced ground state order for this range of $J_2/J_1$, one can expect the system to exhibit RC behavior at temperatures higher than for a single layer.\cite{2} Below we present quantitative results for several experimentally accessible quantities.

We begin by defining spin operators that are symmetric and antisymmetric with respect to interchange of the two layers:

$$\vec{S}_\pm(i) = \vec{S}_{1i} \pm \vec{S}_{2i}.$$  \hspace{1cm} (2)
Using these we have calculated the correlation functions

\[ C_{\pm}(\vec{r}_i - \vec{r}_j) = \langle S_{\pm}^z(i) S_{\pm}^z(j) \rangle, \] (3)

the corresponding static structure factors

\[ S_{\pm}(\vec{q}) = \frac{1}{2L^2} \sum_{i,j} e^{i\vec{q} \cdot (\vec{r}_i - \vec{r}_j)} C_{\pm}(\vec{r}_i - \vec{r}_j), \] (4)

and static susceptibilities

\[ \chi_{\pm}(\vec{q}) = \frac{1}{2L^2} \sum_{i,j} e^{i\vec{q} \cdot (\vec{r}_i - \vec{r}_j)} \int_0^\beta d\tau \langle S_{\pm}^z(i, \tau) S_{\pm}^z(j, 0) \rangle, \] (5)

where \( S_{\pm}^z(i, \tau) = e^{\tau \hat{H}} S_{\pm}^z(i) e^{-\tau \hat{H}} \), and \( L \) is the linear size of the system. The normalization in (4) and (5) has been chosen such that the standard definitions for a single plane are recovered for \( J_2/J_1 = 0 \). For the numerical simulations we have used a QMC method based on stochastic series expansion (a generalization of Handscomb’s method), which is free from errors of the “Trotter approximation” used in standard methods. All results presented here are for 2\times64\times64 lattices (\( L = 64 \)).

In order to extract the correlation length we fit the correlation function \( C_-(r) \) to the nonlinear \( \sigma \)-model forms discussed in Ref. 19. The results are shown in Fig. 1. For \( J_2/J_1 = 0 \) we find good agreement with the results by Makivić and Ding. The enhancement of the correlations with increasing \( J_2/J_1 \) is evident. The correlation length quickly approaches the size of the lattice as \( T \) is lowered below \( T/J_1 \approx 0.4 - 0.5 \), and in order to obtain reliable results for \( \xi \) at lower temperatures lattices larger than \( L = 64 \) would have to be used.

The correlation lengths graphed in Fig. 1 exhibit the exponential growth characteristic of the RC regime, as expected. In order to more quantitatively address the question at what temperature the RC description is valid for a given \( J_2/J_1 \) we study the ratio \( S_-(\pi, \pi)/[T \chi_-(\pi, \pi)] \). In a classical system one alway has \( S/(\chi T) = 1 \) (for any \( \vec{q} \)). A characteristic of the quantum antiferromagnet in the RC regime is that this relation remains satisfied close to the antiferromagnetic wave-vector. On the other hand, for a single plane in the QC regime \( S(\pi, \pi)/[T \chi(\pi, \pi)] \approx 1.1 \). For a two-layer model in the QC regime,
the gapped mode cannot be neglected if the temperature is of the same order as the gap, 
\[ \Delta \propto \sqrt{J_1 J_2}, \] and 
\[ S(\pi, \pi)/[T \chi(\pi, \pi)] \] will probably differ slightly from the above one-layer 
\( \sigma \)-model prediction. In any case, the RC value for this quantity should be 1, and is hence 
useful for determining whether the system is in the RC regime or not. The results displayed 
in Fig. 2 clearly indicate that the coupled planes approach the RC regime considerably faster 
than a single plane.

The frequency integrated neutron scattering intensity is given by a combination of the 
structure factors \( S_+ (\vec{q}) \) and \( S_- (\vec{q}) \), with weights depending on the momentum transfer per-
pendicular to the planes. Since the fluctuations corresponding to \( S_+ (\vec{q}) \) are gapped at 
\( \vec{q} = (\pi, \pi) \), \( S_+ (\pi, \pi) \) saturates below a temperature set by the gap, whereas \( S_- (\pi, \pi) \) diverges. Fig. 3 shows \( J_2 / J_1 = 0.1 \) results for \( \vec{q} \) close to \( (\pi, \pi) \). The ratio \( S_- (\pi, \pi) / S_+ (\pi, \pi) \) is \( \approx 4 \) already at \( T / J_1 = 0.5 \), and increases rapidly with decreasing \( T \). At \( T / J_1 = 0.4 \) the ratio is almost 30 (not shown in the figure). The smallness of \( S_+ (\vec{q}) \) for \( \vec{q} \) close to \( (\pi, \pi) \) may make the detection of the high-energy mode difficult, in particular because the even 
and odd modes cannot be completely separated experimentally.

In Fig. 4 shows results for the uniform susceptibility per spin, \( \chi = \chi_+ (q = 0) \). The 
susceptibility for \( J_2 / J_1 \leq 0.2 \) is very close to the single-plane result over the whole tempera-
ture range considered here. The linear behavior seen for the single plane in the temperature 
regime \( 0.35 \lesssim T / J \lesssim 0.55 \) is in close quantitative agreement with the prediction for the QC 
regime, despite of the fact that the system actually crosses over to the RC regime at these 
temperatures (see Fig. 2). An approximately linear behavior persists at these tempera-
tures also for the systems with \( J_2 / J_1 = 0.2 \) and 0.5, which are even deeper inside the RC 
regime. It would be interesting to compare the behavior with the QC and RC predictions for 
these couplings. As discussed above, this probably requires calculations for a full two-layer 
non-linear \( \sigma \)-model.

We now turn to what we consider the most promising experiment for determining \( J_2 \) 
in the insulating regime. Recently, we showed that the NMR rates \( 1/T_1 \) and \( 1/T_2 \) for 
\( \text{La}_2\text{CuO}_4 \) measured by Imai et al. and Matsumura et al. are well reproduced within the
single-layer Heisenberg model and known hyperfine form factors (for the high-temperature regime, similar results were obtained by Sokol and co-workers). At high temperatures both $1/T_1$ and $1/T_{2G}$ show evidence of QC behavior, although the proximity to the RC and LM regimes influences the behavior as well. As shown above, even a small intrabilayer coupling pushes the system considerably further inside the RC regime. As a consequence, a system with $J_2/J_1 \approx 0.1 - 0.2$ should exhibit no QC behavior in the experimentally accessible temperature range. Below we present QMC results for $1/T_{2G}$ which should be useful for direct comparisons with experiments.

The gaussian component of the spin-echo decay rate is related to the nuclear spin-spin interactions mediated by the electronic spins. The coupling of a $^{63}$Cu nuclear spin $\vec{I}_0$ at site 0 in plane $a$ to surrounding electronic spins is approximately given by the Mila-Rice form:

$$\hat{H} = \sum_{\delta} \langle \vec{I}_0 \cdot \vec{S}_{a\delta} \rangle,$$

where $\delta$ denotes a nearest-neighbor of site 0. The hyperfine coupling constants $A_{\perp}, A_{\parallel}$ and $B$ are known from Knight shift measurements. Pennington and Slichter derived the following form for $1/T_{2G}$, expected to be valid for the cuprates:

$$\frac{1}{T_{2G}} = \left[ \frac{0.69}{2\hbar^2} \sum_{\vec{x} \neq 0} J_z^2(0, \vec{x}) \right]^{1/2}.$$  (7)

Here $J_z(\vec{x}_1, \vec{x}_2)$ is the $z$-component of the induced interaction between nuclei at $\vec{x}_1$ and $\vec{x}_2$,

$$J_z(\vec{x}_1, \vec{x}_2) = -\frac{1}{2} \sum_{i,j} A(\vec{x}_1 - \vec{r}_i) A(\vec{x}_2 - \vec{r}_j) \chi(i-j),$$  (8)

where for the hyperfine coupling (8) one has $A(0) = A_{\parallel}$, $A(1) = B$, and $A(r) = 0$ otherwise. $\chi(i-j)$ is the static susceptibility written in coordinate space. Note that $\vec{x}$ in Eq. (7) runs over all the spins of both planes, except the spin at site 0 in the plane where the nucleus considered resides. The constant 0.69 in (7) is the natural abundance of the $^{63}$Cu isotope.

In Fig. 5 we present results for $J_1/T_{2G}$ versus $T/J_1$ in units of K/s. The results expected in an experiment can be obtained by dividing with the relevant value of $J_1$. We have
used the standard experimental values for the hyperfine couplings; \( B = 41 \text{kOe}/\mu_B \) and 
\( A_{||} = -4B \). In Ref. 18 we have shown that the QMC results for \( J_2 = 0 \) agree well with the 
measurements on \( \text{La}_2\text{CuO}_4 \) (the best agreement is obtained with a slightly smaller value 
for \( B; B \approx 37 \text{kOe}/\mu_B \)). We believe that our \( 1/T_{2G} \) QMC results in Fig. 5 will be useful for 
determining the value of \( J_2 \) in \( \text{YBa}_2\text{Cu}_3\text{O}_6 \), provided that measurements can be carried out 
in the regime of temperatures \( 500 \text{K} \lesssim T \lesssim 800 \text{K} \). A potential difficulty is that \( J_1 \) has to be 
known to rather high accuracy in order to establish the relation to the temperature scale of 
Fig. 5.

In summary, we have studied the effects of a small intrabilayer coupling on the properties 
of the two-dimensional Heisenberg model. Even coupling ratios \( J_2/J_1 \) as small as 0.1-0.2 push 
the boundary of the RC regime up close to the highest temperatures accessible experimentally. 
This should have detectable consequences for a number of quantities. In particular, 
we suggest that measurements of the spin-echo decay rate \( 1/T_{2G} \) at high temperatures would 
be useful for determining the strength of the intrabilayer coupling in \( \text{YBa}_2\text{Cu}_3\text{O}_6 \). The spin-
lattice relaxation rate, which we have not yet calculated for the two-layer model, should also 
be a sensitive probe.

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FIGURES

FIG. 1. The spin-spin correlation length vs. temperature for various strengths of the intrabilayer coupling.

FIG. 2. The ratio $S_-(\pi, \pi)/[T \chi_-(\pi, \pi)]$ vs. $T$ for various intrabilayer couplings. In the RC regime this ratio is 1. The dashed line is the nonlinear $\sigma$-model result for a single plane.

FIG. 3. The odd (upper panel) and even (lower panel) static structure factors for $J_2/J_1 = 0.1$ close to the antiferromagnetic wave-vector $\vec{q} = (\pi, \pi)$ at temperatures close to $T/J_1 = 0.5$.

FIG. 4. The temperature dependence of the uniform susceptibility per spin for various intrabilayer coupling strengths.

FIG. 5. The $^{63}$Cu spin-echo decay rate $1/T_{2e}$ multiplied by the in-plane coupling $J_1$ vs. $T/J_1$ for various values of the intrabilayer coupling strength.
$J_2/J_1 = 0.0$

$J_2/J_1 = 0.1$

$J_2/J_1 = 0.2$

$J_2/J_1 = 0.5$

FIG. 1
T/J

$S(\pi,\pi) / [T\chi(\pi,\pi)]$

$J_2 / J_1 = 0.0$
$J_2 / J_1 = 0.1$
$J_2 / J_1 = 0.2$
$J_2 / J_1 = 0.5$

FIG. 2
FIG. 3
\[ \chi = \frac{J_2}{J_1} \]

FIG. 4
