On the Bilayer Coupling in the Yttrium-Barium Family of High Temperature Superconductors

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Abstract

We present and solve a model for the susceptibility of two CuO$_2$ planes coupled by an interplane coupling $J_\perp$ and use the results to analyze a recent "cross-relaxation" NMR experiment on Y$_2$Ba$_4$Cu$_7$O$_{15}$. We deduce that in this material the product of $J_\perp$ and the maximum value of the in-plane susceptibility $\chi_{\text{max}}$ varies from approximately 0.2 at $T = 200$ K to 0.4 at $T = 120$ K and that this implies the existence of a temperature dependent in-plane spin correlation length. Using estimates of $\chi_{\text{max}}$ from the literature we find $5 \text{ meV} < J_\perp < 20 \text{ meV}$. We discuss the relation of the NMR results to neutron scattering results which have been claimed to imply that in YBa$_2$Cu$_3$O$_{6+x}$ the two planes of a bilayer are perfectly anticorrelated. We also propose that the recently observed 41 meV excitation in YBa$_2$Cu$_3$O$_7$ is an exciton pulled down below the superconducting gap by $J_\perp$. 
In the yttrium-barium (Y-Ba) family of high temperature superconductors the basic structural unit is a "bilayer", consisting of two CuO$_2$ planes; the bilayers are separated by CuO chains. Neutron scattering \[1\] and more recently NMR experiments \[2\] have shown that the Cu spins on adjacent planes in a bilayer are coupled. Intra-bilayer coupling has been shown theoretically to lead to a "spin gap" \[3–6\] similar to that observed \[7\] in NMR experiments on underdoped members of the YBa family. In view of the great importance of the spin gap phenomenon, a quantitative analysis of the spin dynamics of a bilayer is desirable. In this letter we provide this analysis and use the results to interpret NMR and neutron scattering experiments.

Focus on the two planes of a bilayer, and neglect coupling to other bilayers. We label the spin degrees of freedom by an index $a = 1, 2$ distinguishing planes, and a site index $i$. We then define the susceptibility
\[
\chi^{ab}(q, \omega) = \int_0^\infty dt \, e^{i\omega t + \vec{q} \cdot (\vec{R}_i - \vec{R}_j)} \left\langle \left[ S^a_i(t), S^b_j(0) \right] \right\rangle.
\] (1)

Because of the symmetry under exchange of planes, $\chi^{ab}$ has only two independent components, $\chi^{11}(q, \omega) = \chi^{22}(q, \omega)$ and $\chi^{12}(q, \omega) = \chi^{21}(q, \omega)$. The two independent components of $\chi$ may be taken to be the even and odd (under the interchange of planes) components $\chi\text{even,odd} = \chi_{11} \pm \chi_{12}$. In a system with antiferromagnetic coupling one expects that at large wavevectors the odd-parity spin fluctuations are softer than the even parity spin fluctuations.

Further analysis requires a model. We shall assume that the interplane coupling $J_\perp$ is weak in comparison to the energies determining the spin susceptibility $\chi_0(q, \omega)$ of a single plane and that its effects may be modeled via the RPA. Thus we write
\[
\chi^{-1} = \begin{bmatrix} \chi_0^{-1}(q, \omega) & -J_\perp \\ -J_\perp & \chi_0^{-1}(q, \omega) \end{bmatrix}.
\] (2)

We further assume $\chi_0(q, \omega)$ has the scaling form $\chi_0(q, \omega) = \chi_0 \xi^z f(q\xi, \omega/\xi^z)$, where $f$ is a scaling function normalized so that $f(0, 0) = 1$, $z$ is the dynamical exponent, $\xi$ is a correlation length and $\vec{q}$ is measured from an ordering wavevector $\vec{Q}$ which for the present discussion is arbitrary. $\vec{Q}$ is believed to be of the order of $(\pi, \pi)$ in high $T_c$ materials. From Eq. \[2\] we see
that $\chi^{11}(q, \omega) = \chi_0(q, \omega)/(1 - (J_{\perp} \chi_0(q, \omega))^2)$ and $\chi^{12}(q, \omega) = J_{\perp} \chi_0(q, \omega)^2/(1 - (J_{\perp} \chi_0(q, \omega))^2)$.

The crucial parameter controlling the susceptibilities in the static limit is

$$
\Delta = J_{\perp} \chi_{\text{max}} = J_{\perp} \chi_0 \xi^2.
$$

We must assume $\Delta < 1$ so that the material has no long range order. If $\Delta^2 \ll 1$ then $\chi^{11} \approx \chi_0$ and $\chi^{12} \approx J_{\perp} \chi_0^2$. In this limit the interplane coupling has a weak effect and the RPA is an appropriate model. On the other hand, if $\Delta^2 \approx 1$ then the interplane coupling is strong and the use of the RPA may be questioned.

We now turn to the NMR experiments of interest. These are $T_2$ experiments performed on $Y_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$, a material in which the single-chain structure of $\text{YBa}_2\text{Cu}_3\text{O}_7$ alternates with the double-chain structure of $\text{YBa}_2\text{Cu}_4\text{O}_8$ \[8\]. As a result, atoms on different planes of a bilayer have somewhat different local environments and therefore somewhat different NMR resonance frequencies, which may be independently studied. Despite the differences in local environment the electronic properties of the two planes are not very different \[8\], so it is still appropriate to model the electronic properties with Eqs. (1-5). Now the NMR $T_2$ measures the rate at which a nuclear spin is depolarized by interacting with other nuclear spins, i.e. it measures the nuclear-spin–nuclear-spin interaction strength. In high $T_c$ materials the dominant contribution to the nuclear-spin nuclear-spin interaction comes from polarization of electronic spins, and may be related to the static limit of the real part of the electronic spin susceptibility. In $Y_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$ it is possible to measure $T_2$, the rate at which a spin in one plane is depolarized by spins in the same plane, and $T_{2\perp}$, the rate at which a spin in one plane is depolarized by spins in the other plane. $T_2$ is related to the electronic spin susceptibility by \[9\]

$$
\frac{1}{T_2} = \left[ \sum_q \left[ A^2_{q \chi_{\perp}} \right] - \left( \sum_q A^2_{q \chi_{\perp}} \right)^2 \right]^{1/2},
$$

while $T_{2\perp}$ is given by \[10\]

$$
\frac{1}{T_{2\perp}} = \left[ \sum_q \left[ A^2_{q \chi_{\perp}} \right] \right]^{1/2}.
$$

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We have calculated $T_2$ and $T_{2\perp}$ from Eqs. (2-4-5). The precise values obtained depend upon the form chosen for $f(q\xi)$. We have used two forms for $f(x) = f(q\xi, \omega = 0)$: a Lorentzian, $f(x) = 1/(1 + x^2)$, and a Gaussian, $f(x) = \exp(-\log(2)x^2)$ (the $\log(2)$ is introduced so $f(x = 1) = 1/2$). We measure $\xi$ in units of the lattice constant, we set $\hbar = 1$ and assume that the hyperfine coupling can be approximated by its value at $Q, A_Q$. We find

$$\frac{1}{T_2} = A_Q^2\chi_0\xi g_{in}(\Delta)$$
(6)

and

$$\frac{1}{T_{2\perp}} = A_Q^2J_\perp\chi_0^2\xi^2 g_{\perp}(\Delta)$$
(7)

where $g_{in}$ and $g_{\perp}$ are defined in terms of the function $f(x)$ via

$$g_{in}^2(\Delta) = \frac{1}{2\pi} \left[ \int_0^\infty dx \frac{f^2}{(1 - \Delta^2 f^2)^2} - \frac{1}{2\pi\xi^2} \left( \int_0^\infty dx \frac{f^2}{1 - \Delta^2 f^2} \right)^2 \right]$$
(8)

$$g_{\perp}^2(\Delta) = \frac{1}{2\pi} \int_0^\infty dx \frac{f^4}{(1 - \Delta^2 f^2)^2}.$$  
(9)

In writing Eqs. (8,9) we have assumed that the correlation length is so long that lattice effects may be neglected. We have investigated this issue by performing the exact integrals numerically. The parameter governing the size of the lattice effects is $(\pi\xi)^{-1}$; for $\xi \geq 1$ we have found that they are negligible.

In Fig. 1 we present the calculated results for $T_2/T_{2\perp} = \Delta g_{\perp}(\Delta)/g_{in}(\Delta)$. From the experimental values $T_2/T_{2\perp} = 0.15$ at $T = 200$ K and $T_2/T_{2\perp} = 0.30$ at $T = 120$ K [2] we obtain $\Delta \approx 0.2$ at $T = 200$ K and $\Delta \approx 0.4$ at $T = 120$ K. Note that even at the lowest temperature we find that the system is in the small $\Delta$ regime in which $T_2/T_{2\perp}$ is linear in $\Delta$, suggesting that the between-planes coupling is sufficiently small that the RPA formula is justified.

Now $Y_{2}Ba_{4}Cu_{7}O_{15}$ has a doping somewhere between $YBa_{2}Cu_{3}O_{7}$ and $YBa_{2}Cu_{3}O_{6.7}$ [3]. It seems clear that the strength of the magnetic correlations is a relatively rapid function
of doping and increases as one moves from $O_7$ to the insulator, while $J_\perp$ is unlikely to be a sensitive function of doping. It therefore seems very likely that for dopings corresponding to $\text{YBa}_2\text{Cu}_3\text{O}_{6.7}$ and below, $\chi_{\text{max}}$ is so large that $\Delta \approx 1$ and the planes are so strongly coupled that the properties are not linear in $J_\perp$.

The magnitude of $J_\perp$ may be determined if $\chi_{\text{max}}$ is known and conversely. If the susceptibility were only weakly $q$-dependent then the measured uniform susceptibility $\chi_{\text{uniform}} \approx 2 \text{ states/eV-Cu}$ [11] would provide a good estimate for $\chi_{\text{max}}$ and our value $\Delta \approx 0.4$ would imply $J_\perp \approx 0.2$ eV. Such a value is very difficult to justify on microscopic grounds because the insulating antiferromagnetic parent compounds of the high $T_c$ superconductors have in-plane exchange constants $J_{\text{in-plane}} \approx 0.12$ eV and it is generally believed that $J_\perp \ll J_{\text{in-plane}}$. Therefore, we believe the cross-relaxation results imply $\chi_{\text{max}} \gg \chi_{\text{uniform}}$. A similar conclusion has been drawn from an analysis of the magnitude of the in-plane $T_2$ measured on $\text{YBa}_2\text{Cu}_3\text{O}_7$ [9], combined with various assumptions about magnitudes of hyperfine couplings. The magnitude of the hyperfine coupling drops out of the present analysis. The estimate $J_\perp \approx 10 - 20$ meV has been obtained from band structure calculations [12], implying $\chi_{\text{max}} \approx 40 \text{ states/eV-Cu}$. The in-plane $T_2$ experiment led to the value $\chi_{\text{max}} \approx 80 \text{ states/eV-Cu}$ [4] implying $J_\perp \approx 5$ meV.

In summary, the cross-relaxation experiment shows that the real part of the susceptibility at some non-zero $q$ is much larger than the uniform susceptibility. Now the temperature dependence of the $T_2$ rates must be due to the temperature dependence of this antiferromagnetic maximum. Two scenarios have been proposed for the temperature dependence: in the antiferromagnetic scenario the temperature dependent quantity is the correlation length $\xi$. In the generalized marginal fermi liquid scenario the temperature dependent quantity is the overall amplitude $\tilde{\chi}$ [13]. From Eqs. (6,7) we see that in the regime where $T_{2\perp}$ is linear in $\Delta$ the antiferromagnetic scenario predicts $T_2^3/T_{2\perp}$ is temperature independent, while the marginal fermi liquid scenario predicts $T_2^3/T_{2\perp}$ is temperature independent. The experimentally determined ratios are plotted in Fig. 2 and are more consistent with the antiferromagnetic scenario.
The imaginary parts of the two independent susceptibilities \( \chi_{\text{even}} \) and \( \chi_{\text{odd}} \) are measurable via neutron scattering because they have different dependences on \( q_z \), the momentum transverse to the CuO\(_2\) planes \[14\]. Neutron scattering experiments have been performed on a variety of members of the yttrium-barium family of high-\( T_c \) materials \[1,14–16\]. The experimental result is that only \( \chi_{\text{odd}} \) is seen. At frequencies less than 30 meV and temperatures less than room temperature the even parity fluctuations are claimed to be completely frozen out. The theory of neutron scattering in high \( T_c \) materials is presently controversial. There is no generally accepted model which correctly accounts for the observed lineshapes and temperature dependences. To investigate the connection between the cross-relaxation experiments and neutron scattering we have chosen to calculate the ratio of the \( q \)-integrated even and odd parity susceptibilities. This ratio is insensitive to the precise details of the susceptibilities. For definiteness we used the "MMP", dynamical exponent \( z = 2 \) ansatz

\[
\chi_0(q, \omega) = \frac{\bar{\chi}}{(\xi^{-2} + q^2 - i\omega/\Gamma)}.
\]

Here \( \Gamma \) is a microscopic spin relaxation time. The results depend on \( \Delta \) and on \( \omega_{SF} = \Gamma/\xi^2 \), which is the softest spin fluctuation frequency of a single plane. Of course \( J_\perp \) will reduce this frequency for the odd parity channel and increase it for the even channel. Results are shown in Fig. 3 for several values of \( \Delta \). We see that the relative weight of the even parity fluctuations becomes small only for \( \Delta > 0.5 \). We believe that the neutron results, which seem to require a \( \Delta > 0.5 \), are not in contradiction to our analysis of the cross-relaxation experiment, which yielded a \( \Delta \leq 0.4 \), because the strongest neutron evidence for locked bilayers was obtained from a study of YBa\(_2\)Cu\(_3\)O\(_{6.5}\) \[1\], which as we have previously noted is closer to the magnetic instability than Y\(_2\)Ba\(_4\)Cu\(_7\)O\(_{15}\), and therefore may be expected to have a larger \( \Delta \).

Another experiment in which the even parity fluctuations were not seen at all was an observation of a rather sharp peak at an energy of 41 meV in the superconducting state of YBa\(_2\)Cu\(_3\)O\(_7\). In this material there is some evidence of a spin fluctuation peak in the normal state at a similar energy, but it is much broader in \( q \) and \( \omega \). We suggest that the 41 meV peak is an exciton pulled down below the superconducting gap edge by the interplane coupling \( J_\perp \). We note that YBa\(_2\)Cu\(_3\)O\(_7\) has somewhat weaker magnetic correlations than
$Y_2\text{Ba}_4\text{Cu}_7\text{O}_{15}$, so one would expect $\Delta < 0.4$ in the normal state. Now in the superconducting state, the presence of the gap implies that $\chi''$ becomes very small at energies less than the gap, and has a peak (the details of which depend in a complicated way on the details of the superconducting order parameter) at the gap edge. This behavior of $\chi''$ implies via the Kramers-Kronig relation a large enhancement of the real part, $\chi'$, at the gap edge. We propose that this enhanced $\chi'$ leads to a weakly damped pole, at a frequency of order the gap edge, in the odd parity channel of the RPA formula, Eq. [2], and that this pole produces the feature seen experimentally. Our proposal provides a natural explanation for the sharpness of the feature and for the nearly perfect bilayer correlation observed at low temperatures. If this scenario is correct the even parity component of the neutron scattering signal should appear above $T_c$.

In summary, our analysis of the cross-relaxation experiments suggests that the between-planes coupling $J_\perp$ is small, but has non-negligible effects which furthermore increase as the temperature is decreased. These are precisely the assumptions made in the theories which attribute spin gap formation to interplane pairing, so we believe our results tend to support this picture.

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REFERENCES

[1] J. M. Tranquada, G. Shirane, B. Keimer, S. Shamoto and M. Sato, Phys. Rev. **B40**, 4503 (1989) and Phys. Rev. **46**, 5561 (1992).

[2] R. Stern, M. Mali, J. Roos, D. Brinkmann, unpublished.

[3] B. L. Altshuler and L. B. Ioffe, Solid State Communications **82** 253 (1992).

[4] A. J. Millis and H. Monien, Phys. Rev. Lett. **70**, 2813 (1993).

[5] B. L. Altshuler, L. B. Ioffe, A. I. Larkin and A. J. Millis, JETP Lett. **59**, 65 (1994) and unpublished.

[6] M. Ubbens and P. A. Lee, Phys. Rev. **B50**, 438 (1994).

[7] M. Takigawa et al., Phys. Rev. **B42**, 243 (1991).

[8] R. Stern, M. Mali, I. Mangelschots, J. Roos, D. Brinkmann, J-Y. Genoud, T. Graf and J. Muller, Phys. Rev. **B50**, 426 (1994).

[9] C. H. Pennington and C. P. Slichter, Phys. Rev. Lett. **66**, 381 (1991).

[10] H. Monien and T. M. Rice, Physica **C235-240**, 1705 (1994).

[11] R. E. Walstedt et. al., Phys. Rev. **B45**, 8074, (1992).

[12] O. K. Andersen et. al. Phys. Rev. **B49** 4145 (1994).

[13] Q. M. Si, Int. J. of Mod. Phys. **B8**, 47 (1994).

[14] J. M. Tranquada et al., Phys. Rev. **B40**, 4503 (1989).

[15] J. Rossat-Mignod et al., Physica **C185**, 86 (1991).

[16] H. A. Mook, M. Yethraj, G. Aeppli, T. E. Mason and T. Armstrong, Phys. Rev. Lett. **70** 3490 (1993).
FIG. 1. Ratio of cross-relaxation rate $1/T_{2\perp}$ to in-plane relaxation rate $1/T_2$ plotted versus coupling parameter $\Delta = J_{\perp} \chi_{\max}$ for Lorentzian (dotted line) and Gaussian (solid line) form factors and calculated from Eqs. (6-9).
FIG. 2. Experimentally determined ratio of $1/T_{2\perp}$ to $n$th power of $1/T_2$ for $n=1$ (□), 2 (●), 3 (○) in arbitrary units. That the $n=3$ (○) curve has less temperature dependence than the $n=2$ (●) curve suggests the existence of a growing magnetic correlation length.
FIG. 3. Calculated ratio of $q$-integrated odd-parity neutron absorption to $q$-integrated even parity neutron absorption, plotted versus frequency for $\Delta = J_\perp \chi_{max} = 0.0, 0.1 \ldots 0.9$. $\Delta = 0.0$ corresponds to the lowest curve and $\Delta = 0.9$ to the top curve.