Remarks on Spin Gaps and Neutron Peak Selection Rules in YBCO — Do Interlayer Tunneling and Interlayer RVB as Mechanisms for Cuprate Superconductors Differ?

Philip W. Anderson

Joseph Henry Laboratories of Physics, Princeton University, Princeton, N.J. 08544

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Abstract

We point out that both superexchange between $CuO_2$ layers, and interlayer tunneling, derive from frustrated one-particle hopping between layers, and that they should be treated on an equal footing. Doing so, we arrive at a new view of the nature of pairing in the cuprate superconductors, which explains the striking even ↔ odd selection rule observed by Keimer in neutron scattering by YBCO.
Recently, two different hypotheses have been put forward for the “spin gap” phenomenon exhibited by the bilayer structures of cuprate superconductors. (As in YBCO and BISCO 2-layer materials). Millis and Monien and co-workers [1] have proposed that what is occurring is pair formation between the two layers with one of the pair on each layer, motivated by the interlayer antiferromagnetic superexchange. Strong and myself [2] have proposed that the interlayer pair tunneling Hamiltonian produces a correlated state with pairing on each layer for each momentum \( k \), but the different \( \vec{k} \)'s are independently phased. Both postulate “preformed BCS pair” states as the essential nature of the spin gap phenomenon.

Although it is not explicitly mentioned in the former paper, both theories have one vital element in common: the coherent motion of single electrons between the two layers must be blocked. As I have emphasized elsewhere, [3] antiferromagnetic “superexchange” interaction exists only if the single-particle kinetic energy is frustrated, and is a consequence of the second-order, virtual action of this kinetic energy, causing hopping of electrons between the relevant sites. If the kinetic energy were diagonalized, the Fermi surface would be split into separate surfaces for even and odd linear combinations of the two layers, which, being orthogonal, would exhibit only ferromagnetic exchange interactions. (The reader should note that although the rest of this article will be couched in language referring only to bilayer materials, only a slight modification allows one to present similar arguments for coupled layers either single or multiple layers.)

In fact, the Millis-Monien et al papers miss another feature of interlayer superexchange. The hopping matrix element \( t_\perp \) between layers is of course diagonal in \( k_{||} \), the interlayer momentum, so that superexchange, \( \sim t_\perp^2/U \), must also contain an extra momentum \( \delta \)-function. That is,

\[
\mathcal{H}_{SE} = \sum_k \sum_{k'} \left( c_{k\uparrow}^{(1)} c_{k'\downarrow}^{\dagger(2)} c_{k'\downarrow} c_{k\uparrow}^{(1)} \right) \times t_k \cdot t_{k'} / \text{energy denom}
\]

and the only way to satisfy the pairing condition is for \( k' = -k \). Thus these two mechanisms share the characteristic \( k \)-diagonality, which leads to approximate independence of different
parts of the Fermi surface; and the Strong-Anderson mechanism for producing a spin gap is common to both.

A third relevant theory paper is the weak-coupling solution of the two-chain Hubbard model by Balents and Fisher [4], in which much of the relevant region of parameter space seems to exhibit the same fixed point with a spin gap and one gapless charged excitation, which seems to interpolate between the pairing schemes of Ref. (1) and (2).

We argue here that the two mechanisms for spin gaps are not incompatible but complementary. They result from the same phenomenon of virtual hopping between layers, which is of course only virtual because the direct coherent hopping at zero frequency is blocked by interaction effects. Both the spin gap phenomenon and superconductivity itself are explicable on the same basis.

We may understand the superexchange phenomenon in Mott insulators by realizing that the orbitals of opposite-spin electrons need not be orthogonal. Thus a possible path for exchange of two electrons of opposite spins is for the second electron (of down spin, say) to hop back from site 2 to site 1, during the period while the electron of up spin, while nominally on site 1, has made a virtual transition to site 2. Thus if \( a^\dagger_1 \sigma \) and \( a^\dagger_2 \sigma \) are the properly orthogonalized one-electron operators for the two sites, the actual one-electron operators are

\[
c^\dagger_1 = \frac{(a^\dagger + \epsilon a^\dagger_2)}{\sqrt{1 + \epsilon^2}}
\]

\[
c^\dagger_2 = \frac{(a^\dagger_2 + \epsilon a^\dagger)}{\sqrt{1 + \epsilon^2}}
\]

where

\[
\epsilon = t/U.
\]

These are not orthogonal because they need not be if they belong to opposite-spin electrons. The superexchange energy is then

\[
J = \epsilon t.
\]
This picture can be renormalized, as discussed in Herring [5], by taking into account
interactions between the two electrons along the exchange path, but the physics remains the
same.

\( J \) is the amplitude for interchange of spins keeping the same charge state. We may also
ask for the pair tunneling amplitude, i.e., the amplitude for an up-spin down-spin pair to
tunnel from one site to another. This is of course charge disfavored for atomic sites, but
for tunneling between metallic layers there is no charge rigidity. In this case the down-spin
electron hops in the opposite direction, and the amplitude is also

\[ E_J = t_\perp \epsilon , \]

with one hop taking place by virtue of non-orthogonality of wave functions in the two layers,
the second by the actual frustrated one-electron matrix element. This two-particle tunneling
process is the basis of the interlayer theory. It, like superexchange, can be described by an
effective Hamiltonian for low energy states.

It becomes clear that there is no sharp way to distinguish between antiferromagnetic
superexchange and pair tunneling mechanisms for spin gaps and superconductivity if we
look at the order parameters which can result.

First let us set up some notation. Let \( k \) be a momentum near the Fermi momentum of
the two chains or planes, 1 and 2. Orthonormal electron operators are defined as

\[
\begin{align*}
a_{1\uparrow}^{(1)+} &= a_1^+ \\
a_{2\uparrow}^{(2)+} &= a_2^+ \\
a_{-1\downarrow}^{(1)+} &= a_{-1}^+ \\
a_{-2\downarrow}^{(2)+} &= a_{-2}^+ 
\end{align*}
\]

and these can combine into the even and odd eigen-operators of the kinetic energy,

\[
\begin{align*}
a_e^+ &= \frac{a_1^+ + a_2^+}{\sqrt{2}} \quad \text{etc.} \\
a_o^+ &= \frac{a_1^+ - a_2^+}{\sqrt{2}} \quad \text{etc.}
\end{align*}
\]

We know that the kinetic energy operator may be written
\[ \mathcal{H}_k = \epsilon_k (n_{k_1} + n_{k_2}) \]  
\[ + t_{\perp} (a_1^+ a_2 + a_{\perp 1}^+ a_{\perp 2}) \]
\[ = (\epsilon_k + t_{\perp}) (a_o^+ a_o + a_{\perp o}^+ a_{\perp o}) \]
\[ + (\epsilon_k - t_{\perp}) (a_e^+ a_e + a_{\perp e}^+ a_{\perp e}) \]

but in the case where interactions within the layers or chains are sufficiently strong, the splitting given by the last expression into even and odd eigenstates is not expressed in the actual state: \( t_{\perp} \) causes primarily virtual transitions \( \text{[6]} \), as in the Mott insulator. (More precisely: \( t_{\perp} \) causes no coherent transitions). This is an experimental fact in several of the cuprates, according to photoemission and infrared spectroscopy. \( \text{[7,8]} \). Instead of splitting, the electrons hop virtually, so that the electron operators which describe the actual eigenexcitations are roughly

\[ c_1^+ = \frac{(a_1^+ + \epsilon a_2^+)}{\sqrt{1 + \epsilon^2}} \]

\[ c_2^+ = \frac{(a_2^+ + \epsilon a_1^+)}{\sqrt{1 + \epsilon^2}} \]

Now let us imagine that by the pair tunneling argument we have derived an order parameter in which the electrons are paired in their separate layers:

\[ (\text{O.P.)}_{\text{TL}} = \langle c_1^+ c_{-1}^+ + c_2^+ c_{-2}^+ \rangle \]

\[ = \frac{\langle (a_1^+ a_{\perp 1} + a_2^+ a_{\perp 2}) \rangle + \epsilon (a_{1-}^+ a_{\perp 2} - a_{2-}^+ a_{\perp 1})}{\sqrt{1 + \epsilon^2}} \]

This order parameter is, in terms of orthonormal states, a mixture of the Strong and the Millis pairings; it may also be written

\[ (\text{OP})_{\text{IL}} = \frac{\langle a_e^+ a_{\perp e} + a_o^+ a_{\perp o} \rangle}{\sqrt{1 + \epsilon^2}} \]

\[ + \frac{\epsilon}{\sqrt{1 + \epsilon^2}} \frac{\langle (a_e^+ a_{\perp e} - a_o^+ a_{\perp o}) \rangle}{\sqrt{1 + \epsilon^2}} \]

\[ = \langle a_e^+ a_{\perp e} \rangle \left( \frac{1 + \epsilon}{\sqrt{1 + \epsilon^2}} \right) + \langle a_o^+ a_{\perp o} \rangle \left( \frac{1 - \epsilon}{\sqrt{1 + \epsilon^2}} \right) \]

Equally, we may imagine a Millis-Monien pairing
\[
\langle c_1^+ c_{-2}^+ + c_2^+ c_{-1}^+ \rangle = \frac{\langle (a_1^+ a_{-2}^+ + a_2^+ a_{-1}^+) \rangle + \epsilon \langle (a_1^+ a_{-1}^+ + a_2^+ a_{-2}^+) \rangle}{\sqrt{1 + \epsilon^2}} 
\]

(7)

Again, in terms of orthogonal orbitals this is a mixture of the two pairings. In terms of even and odd

\[
(O.P.)_{\text{MM}} = \langle a_1^+ a_{-2}^+ \rangle \left( \frac{1 + \epsilon}{\sqrt{1 + \epsilon^2}} \right) - \langle a_2^+ a_{-1}^+ \rangle \left( \frac{1 - \epsilon}{\sqrt{1 + \epsilon}} \right) 
\]

(8)

(Incidentally, neither pairing is of the “s, \(-s\)” type suggested by Scalapino and Yakovenko). The M-M pairing will also lead to superconductivity because of the \(\epsilon\) term which represents interlayer pair tunneling, if there is coupling to other layers as well as to other momenta, through conventional short-range interactions.

These two pairings have the interesting feature that they reinforce each other for the even-even pairing, but have opposite sign for the odd-odd pairing. We suggest here that the effective pairing Hamiltonians coming from the two sources have approximately the same coefficient so that in effect we have only even-even pairing: the order parameter is

\[
\text{O.P.} \simeq \langle a_1^+ a_{-2}^+ \rangle ,
\]

since the odd-odd pairing is favored with \(-\) sign by the superexchange term and favored with \(+\) sign by the interlayer term approximately equally.

This pairing is that appropriate to explain the selection rules observed by Keimer et al [9] in the scattering of neutrons by the bilayer material \(YBa_2Cu_3O_7\). The observation is that a pronounced peak appears, rather sharp in energy at \(\sim 42\) mev but broader than instrumental resolution in momentum space, near \(\pi, \pi\) in the \(ab\) plane Brillouin zone. The peak appears only below \(T_c\) (magnetic scattering reported above \(T_c\) seems to have been an artifact) and is either simply quasiparticle pair production, or the same somewhat enhanced by excitonic interaction effects. More exotic explanations seem incompatible with the experimental facts, in particular, exotic collective excitations seem to have no reason to appear sharply below \(T_c\), and at an energy so close to the supposed value of \(2\Delta\). The \(k_{||}\)-dependence is very compatible
with the idea that coherence factors forbid magnetic scattering between \( k \)'s with energy gaps which have the same sign, and enhance strongly those at energy gaps of opposite sign, which presumably (using BISCO as a model, and relying on Josephson interference measurements) occur near points \( X \) and \( Y \), separated by \( \pi, \pi \) in the zone. The observed BISCO gap would fit the \( k_{||} \)-dependence well.

The dependence on \( k_{\perp} \) in the \( c \)-direction is remarkable. This is a sinusoidal curve with a period given by the inverse of the interplanar spacing, showing that the scattering changes sign between the two planes. An equivalent statement is that scattering satisfies the selection rule even \( \leftrightarrow \) odd as far as symmetry in the pair of planes is concerned.

A little thought convinces one that this cannot be a coherence factor selection rule. That is, if the coherence factor is large between \( k \)-points \( (0, \pi, 0 \) and \( \pi, 0, \pi \)), the gaps \( \Delta_{0\pi0} = -\Delta_{\pi0\pi} \). But then the sign of \( \Delta_{\pi00} \) must be opposite to either one or the other, leading to a second peak, which is not observed, either at \( \pi, \pi, 0 \) or \( 0, 0, \pi \). We propose that this rule holds because at the energy gap the even state is preferentially occupied, the odd state empty. Then the dominant scattering process for a neutron is even \( \rightarrow \) odd, when the pair of particles created is primarily at the energy gap.

The understanding of how this comes about requires us to go rather deeply into the BCS mechanism and the slight generalization that is necessary in this problem.

In conventional BCS, the effective Hamiltonian for a single pair of states is

\[
H_{\text{one pair}} = (\epsilon_k - \mu)(n_{k\uparrow} + n_{-k\downarrow} - 1) \\
+ \Delta_k c_{k\uparrow}^+ c_{-k\downarrow} + \Delta_k^* c_{-k\uparrow}^+ c_{k\downarrow}^+
\]

An irrelevant constant term has been added to make it clear that \( H \) may be taken such that it has no effect on the subspace \( n_{k\uparrow} + n_{-k\downarrow} = 1 \) of states not satisfying the Schrieffer pairing condition.

In the pseudospin representation, one may write (1) in terms of the Nambu spinors \( \tau \) as

\[
H = (\epsilon_k - \mu) \tau_k^z + \frac{\Delta}{2} \tau_k^+ + \frac{\Delta^*}{2} \tau_k^-
\]
and it can be diagonalized by

\[ \Psi_o = u_k \Psi(\tau_k^z = +1) + v_k \Psi(\tau_k^z = -1) \]  

(11)

with

\[ u_k = \cos \frac{\theta}{2}, v_k = \frac{\sin \theta_k}{2} \]

\[ \tan \theta_k = \frac{\Delta_k}{\epsilon_k - \mu} \]

In the present instance, for a pair of planes we have four Fermions belonging to a given \( k \)-vector, \( c^{+\alpha}_{k\uparrow}, c^{-\alpha}_{k\downarrow} \) with \( \alpha = 1, 2 \) a plane index. Alternatively, we may use \( c^{e,o}_{k\uparrow} \) with

\[ c^{e,o}_{k\uparrow} = \frac{1}{\sqrt{2}} (c^{(1)}_{k\uparrow} \pm c^{(2)}_{k\uparrow}). \]  

(12)

There are two calculations which may be carried out. One is in the spin-gap situation, where we neglect all coupling to other momenta. Then the states of momentum \( k \), as a decoupled subspace, couple through the pair tunneling and superexchange interactions. These are, first, from (1)

\[ \mathcal{H}_{SE} = \sum_k \frac{\lambda_k}{2} (c^{+1}_{k\uparrow} c^{-2}_{-k\downarrow} c^{+2}_{-k\downarrow} c^{1}_{k\uparrow}) \]

\[ + (1 \leftrightarrow 2) + \text{H.C.} \]

and, second, as previously proposed,

\[ \mathcal{H}_{PT} = \sum_k \frac{\lambda_k}{2} (c^{+\alpha}_{k\uparrow} c^{-\alpha}_{-k\downarrow} c^{+(\alpha)}_{-k\downarrow} c^{-(\alpha)}_{k\uparrow}) \]

In a similar way to the BCS case, states not satisfying \( n = 2 \) are annihilated by the interaction. In this case the Hamiltonian is number-conserving overall and the state which diagonalizes the sum of the two interactions is simply

\[ \Psi_{sg} = c^{e+}_{k\uparrow} c^{e-}_{-k\downarrow} \Psi_{vac} \]

and all other states are unaffected by the pairing Hamiltonian. This is the spin gap state, identical to that proposed by Strong and Anderson except that the pairing is in the even
state, while the three other \( N = 2 \) states are either repulsive or not lowered in energy. Otherwise the story is unchanged. In the superconducting state we presume the gap function is correlated by couplings to other nearby momentum states and to other planes. We can model this by assuming that the momentum-conserving \( \delta \)-function is not exact but has a finite width \( \delta k \sim \frac{1}{L} \), (an additional length scale which enters the interlayer theory, whose physical consequences we do not explore here). But \( L \to \infty \) leads to physical nonsense when explored in too great detail.

Note that any electron excited from a spin gap state must leave behind a hole in an even state \( c^{(e)}_k \) and any \( k \) in the spin gap state can only accept an electron in an odd state \( c^{(+o)}_k \). Thus the spin gap states exactly satisfy the Keimer selection rule \( e \to o \) for scattering of electrons by neutrons. This reflects the source of the pairing energy in the kinetic energy term.

\[
\text{K.E.} = t_{\perp}(n_e - n_o).
\]

Also note that although the odd states are nominally unpaired, nonetheless adding or removing an electron in an odd state from a spin gapped \( k \)-value destroys the pairing criterion \( n = 2 \) and costs one unit of pairing energy. Thus there is a gap for all one-electron excitations, even though only the even state is paired.

Now we consider the more conventional BCS-like theory. Here we must require phase-coherence of the pairing among all \( k \)-states, so that the pairing Hamiltonian is no longer number-conserving, and may be treated by the usual mean field theory. But when we transcribe our second-order pairing Hamiltonian into “even” and “odd” language, it turns out to read

\[
-\mathcal{H} = \sum_k \lambda_k \sum_{k',L} \left( c^{e+}_{k\uparrow} c^{e+}_{-k'\downarrow} - c^{o}_{k\uparrow} c^{o}_{-k'\downarrow} \right) \times \left( c^{e}_{-k'\downarrow} c^{e}_{k'\uparrow} - c^{o}_{-k'\downarrow} c^{o}_{k'\uparrow} \right)
\]

Thus it is favorable for pairing to occur in even or odd states but not in both together. The resulting problem is more complicated than BCS; in fact, unlike BCS mean field is not
quite adequate to the solution. Basically, the system goes from 4 states empty

\[ (a) = (n_k^e = n_k^o = o) \] to all full \( (b) \) \( (n_k^e = n_k^o = 2) \)

via even pairing only

\[ (c) \] \( n_k^e = 2, n_k^o = o \)

but continuously, with coherent occupation of \( (a), (b), (c) \). A self-consistent BCS calculation gives us

\[
\Delta_{eff}^{(k)} = \lambda_k \sum_{|k'|=k} \langle \chi^{+e}_{k_k} \chi^{+e}_{-k} - \langle \chi^{o+}_{k_k} c^{o+}_{-k} \rangle \rangle
\]

changing sign and therefore vanishing at \( k = k_F \). [11] What we expect will happen, however, is that at \( k \approx k_F \) there will be phase-correlated fluctuations of \( b_k^+ \) and \( b_{k'}^+ \) as in the spin gap state. Thus the actual energy gap does not actually vanish at any \( k \). How to deal in any exact way with the phase fluctuations of the gap is as yet an unsolved problem. I speculate that the mathematics of gap formation may resemble more nearly that of Bose condensation of preformed pairs than the simple pair condensation of BCS.

If we had only the same-layer pairing Hamiltonian which we have used in the past, i.e.,

\[
\sum_k \lambda_k \sum_{k' - k < L} \left( \chi^{+e}_{k_k} \chi^{+e}_{-k} - \langle \chi^{o+}_{k_k} c^{o+}_{-k} \rangle \right)
\]

we obtain \( \lambda_k \lambda_k^{pr} \approx 1 \) as the effective gap equation, with \( \chi_k = \frac{1}{\epsilon_k} \); this can only be satisfied with \( \epsilon_k^2 \langle \lambda_k^2 \rangle \). The pairing amplitude \( b_k = \Delta_k = \sin \theta_k \).

\[
\cos \theta_k = \frac{\epsilon_k}{\lambda_k} = n_k.
\]

A crude approximation is to assume that this same mathematics occurs twice, once for even pairs and once for odd ones. Basically, \( n_k^o \) goes linearly from 2 to 0 from \( \epsilon_k = -\lambda_k \) to 0; and \( n_k^e \) goes linearly from 2 to 0 from \( \epsilon_n = 0 \) to \( \epsilon_k = +\lambda_k \).

The ratio of amplitudes \( e \to e \) and \( o \to o \) to \( e \leftrightarrow o \) can be calculated using this assumption naively. The ratio of occupation factors is equal to
\[
\frac{\int_{-\lambda}^{\lambda} n_k^o d\epsilon \int_{-\lambda}^{\lambda} d\epsilon (2 - n_k^e) + \int_{-\lambda}^{\lambda} n_k^e d\epsilon \int_{-\lambda}^{\lambda} n_k^e d\epsilon}{\int_{-\lambda}^{\lambda} n_k^e d\epsilon \int_{-\lambda}^{\lambda} (2 - n_k^e) d\epsilon + \int_{-\lambda}^{\lambda} n_k^o \int_{-\lambda}^{\lambda} lambda (2 - n_k^o)} = \frac{5}{3}
\]

This leaves out coherence factors which may considerably enhance the ratio (I estimate by a factor 2). This is still less skewed than the data. The state may resemble more the correlated “spin gap” state than this uncorrelated mean field approximation. The “spin gap” give a ratio of 1:0, and it is unreasonable that the superconducting state should be very much lower.

In a forthcoming paper, in collaboration with S. Chakravarty, we shall show how the amplitude, shape and intensity of the neutron peak follows from the above ideas. Many new complexities are caused by the unique nature of the pairing Hamiltonian in this theory, and we look forward to exploring an unexpectedly rich field of physics.

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REFERENCES

[1] A. Millis and H. Monien, Phys. Rev. Lett., 70, 2810 (1993); also recent preprint.

[2] P.W. Anderson and S.P. Strong, Chinese Journal of Physics, to appear.

[3] P.W. Anderson, Advances in Physics, to appear.

[4] L. Balents and M.P.A. Fisher, preprint.

[5] C. Herring, in Magnetism, G. Rado and H. Suhl eds, Vol. II (64), Vol. IV (1966) Acad.
   Press, New York

[6] D.G. Clarke, S.P. Strong, P.W. Anderson, Phys. Rev. Lett., 74, 4499 (1995)

[7] J.R. Campuzano et al, Phys. Rev. Lett., 76, 1533 (1996)

[8] D. Van der Marel, et al, Physica C 235-240, Pt. II, p. 1145, (1994) & references therein.

[9] H.F. Fong, B. Keimer et al., Phys. Rev. Lett., 75, 316 (1995)

[10] P.W. Anderson, Phys. Rev. 112, 1900 (1958); also see Y. Nambu.

[11] Thus as far as the mean field gap function is concerned, the speculation which I proposed
    in 1991 (P.W. Anderson, Physica C 185–89 11–16 (1991) is roughly confirmed; this
    was followed up by A. Balatsky and E.A. Abrahams, Phys. Rev. B45, 13125 (1992).
    However, the physical picture of a conventional energy gap is more descriptive.