C-Axis Electrodynamics as Evidence for the Interlayer Theory of High $T_c$ Superconductivity

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In the interlayer theory of high temperature superconductivity the interlayer pair tunneling (similar to the Josephson or Lawrence-Doniach) energy is the motivation for superconductivity. This connection requires two experimentally verifiable identities. First, the coherent normal state conductance must be too small relative to the “Josephson” coupling energy, and second, the Josephson coupling energy must be equal to the condensation energy of the superconductor. The first condition is well satisfied in the only case which is relevant, $(LaSr)_2CuO_4$; but the second condition has been questioned. It is satisfied for all dopings in $(La – Sr)_2CuO_4$, and also in optimally doped $Hg(Ba)_2CuO_5$ which was measured recently, but seems to be strongly violated in measurements on single crystals of $Tl_2Ba_2CuO_6$. 
The theory that ascribes the phenomenon of high $T_c$ superconductivity in the cuprates primarily to interlayer coupling \[1\] correlates electromagnetic coupling along the $c$-axis (perpendicular, that is, to the $CuO_2$ planes) with the condensation energy of the superconductor. This correlation, which should be particularly sharp for “one-layer” materials, was proposed and roughly tested against data on $(La - Sr)_2CuO_4$ (“214”) in the original paper, \[2\] and the equations were refined in papers by van der Marel et al \[3\] and Leggett \[4\]. In these latter papers, the apparent failure of the relation in $Tl_2Ba_2CuO_6$ “($Tl \ 2201$”) is emphasized, and new, rather unequivocal measurements of $\lambda_c$, the $c$-axis penetration depth by Moler et al \[5\], confirm this contradiction. However, as I show below, there is quite good agreement in a growing number of other cases: 214 at several different doping levels, \[6\] and, very recently, $Hg$ “1201” cuprate, $HgCa_2CuO_4$. \[7\] It appears then, that the $Tl$ salt is the “odd man out” or perhaps not a true one-layer case; this compound exhibits wide swings in $T_c$ with preparation treatment. Because both the $Tl$ and $Hg$ salts have relatively large $c$-axis spacings and comparable $T_c$’s of around 90 K, the contradiction between the two is particularly striking, and it is only less important to confirm the measurements of Ref. \[7\], preferably by another experimental method.

Additional evidence for a major role for interlayer coupling is the observation of a strong bilayer correlation in neutron scattering in YBCO both in the superconducting state (for optimal doping) \[8\] and in the spin-gap regime, \[9\] which is not explicable in “one-layer” theories but receives a natural explanation in the interlayer theory \[10\]. Thus $Tl \ 2201$ stands out in providing contravening evidence against the theory of \[1\].

The ILT theory is very simple in principle. Electron motion for the cuprates in the $c$-direction is incoherent in the normal state. This is unlike most normal metals, which are Fermi liquids and which exhibit coherent transport in all directions. The interlayer hypothesis is that electron pairing in the superconducting state makes this transport coherent, which is actually observed, and which is responsible for the Josephson-like or Lawrence-Doniach-like superconducting coupling between the layers. In conventional superconductors, the
Lawrence-Doniach coupling replaces coherent transport in the normal state, so that the superconductor gains no relative energy, but in the cuprates, actual experimental observations exclude coherent transport in the normal state, so that the c-axis energy is available as a pairing mechanism. [In my theory [1], the mechanism for blocking coherent transport is the non-Fermi liquid nature of the normal metal state.] Thus superconductivity occurs in connection with a two-dimensional (2D) to 3D crossover; if for some reason, one desires a “quantum critical point” to be associated with high $T_c$, that is its nature.

Our concept is, then, that there are two independent ways of measuring the energy coupling the planes together in the superconductor, each direct. The first is, in analogy with the Josephson energy-current relation, to measure the electromagnetic response to vector potentials along the $c$ axis, either by measuring the $c$-axis penetration depth or the $c$-axis transverse plasma frequency. Because of the correlation referred to above, this plasmon in the cuprates lies, unusually, within the superconducting gap, and is clearly visible as a sharp edge in the reflectivity, followed by a dip.

The second measurement is of the condensation energy of the superconductor. Our postulate is that this is wholly, or almost wholly, due to the $c$-axis coupling, so that it should be equal numerically to the maximum possible value of electromagnetic coupling, when all layers are equivalently coupled—that is, only in “one-layer” superconductors. This is equivalent to the statement that $\xi_c \approx \frac{\xi}{2}$ where $\xi_c$ is the $c$-axis correlation length. As pointed out in [2], this is a maximum possible value for $\xi_c$ or a minimum for $\lambda_c$ for multilayer systems such as $Y B C O$ or $B i_2 S r_2 C a C u_2 O_8$ (“BISCO 2 2 1 2”). The condensation energy may be estimated from $T_c$ or $\Delta$, using BCS expressions, but because I am arguing that BCS does not use the correct form of interaction, it can give no better than an order of magnitude estimate, and it is far better to use specific heat data when available. In particular, the dependence on doping, according to such data, of condensation energy is steeper than that of $T_c^2$ so that for underdoped materials one must be especially careful. $T_c$ is roughly proportional to $x$, while $E_{\text{cond}} \propto x^p$ with $p$ previously estimated as $\sim 3$ to 4. The theory of Lee and Wen [11] gives $p = 3$ but has been seriously questioned [12]; specific heat data are therefore more
convincing, if hard to interpret quantitatively \[13\]. The sharp dependence of \( \lambda_{ab} \) on \( x \) which is predicted by Lee and Wen in \[11\] is observed in \[13\] contrary to the criticisms of \[12\] which uses figures from YBCO where effects of doping are less straightforward. It is clear from such data that condensation energy falls off with doping percentage \( x \) more rapidly than \( T_c^2 \).

The basic formulas are as follows: First, for the electromagnetic theory of an interlayer superconductor, the basic London equation is:

\[
\vec{j} = \frac{1}{\lambda^2} \frac{c}{4\pi} \vec{A}
\]

where \( c \) is the speed of light and \( \vec{A} \) is the vector potential. This is the definition of the penetration depth \( \lambda \). Focusing on \( T \ll T_c \), and ignoring the difference between free energy \( F \) and energy \( E \):

\[
\vec{j} = c \frac{\partial F}{\partial \vec{A}} \simeq c \frac{\partial E}{\partial \vec{A}}
\]

The pairing energy in the interlayer theory comes entirely from the coupling between planes, so that one can take \( E \) to be the condensation energy \( E_b \) and assume the coupling energy has the Josephson form

\[
E_b = -E^0_b \cos \theta
\]

where \( \theta \) is the phase difference between the pairs of planes. In the presence of a vector potential

\[
\nabla \theta = \frac{2e}{\hbar c} A
\]

\[
\theta = \frac{2ed}{\hbar c} A
\]

where \( d \) is the spacing between layers, \( e \) is the charge of an electron, and \( \hbar \) is Planck’s constant divided by \( 2\pi \). Combining Eqs. [2], [3] and [4]:

\[
j = 4c E^0_b \left( \frac{a^2 d^2}{\hbar^2 c^2} \right) A
\]

\[
\lambda_c = \frac{\hbar c}{2ed} \frac{1}{\sqrt{4\pi E^0_b}}.
\]
A nearly equivalent measure of the electromagnetic coupling is the $c$-axis plasma frequency. The dielectric constant $\epsilon$ is given in terms of the $\delta$-function “Drude weight”

$$\omega_p^2 = \frac{c^2}{\lambda^2} \tag{6}$$

By

$$\epsilon = -\frac{\omega_p^2}{\omega_p^2} + \epsilon^0 \tag{7}$$

and the edge occurs where $\epsilon$ changes sign, at

$$\hbar\omega_p^c = \frac{\hbar c}{\sqrt{\epsilon_0\lambda}} \tag{8}$$

$$= \sqrt{\frac{4\pi E_b^0}{\epsilon_0}} \times 2ed$$

In the cases of 214, $\sqrt{\epsilon_0}$ is an actually measured quantity from the normal state reflectivity, because no appreciable Drude weight appears in the normal state, and is $\sim 5 \pm 1$. In the cases of $Tl$ and $Hg$ one-layers, $\epsilon_0$ is not well measured. $\lambda$, however, has no dependence on $\epsilon_0$ and is the measured quantity in both of these cases. Figure 1 shows the measured plasma edge for a series of doping levels in 214.

The thermodynamics of optimally doped YBCO has been thoroughly studied by Loram et al [14] and their estimate for the condensation energy per unit volume is:

$$E_b^0 \ (YBCO) = 3.5 \times 10^6 \text{ erg/cc} \tag{9}$$

(Per unit cell per layer, this is about 3 K, which is not far from the BCS estimate of $\frac{N(O)(kT_c)^2}{2}$, taking $N(O)$ to be $\sim 2$ to 3/eV.)

The binding energy for 214 must be estimated from Loram et al’s curves [13,14] (Figure 2), which also, fortunately, shows several doping levels. For the optimal doping level, 17 to 20%, $E_b$ can be estimated with the identities:

$$\int (c_N - c_s) \,dT = E_b = \int T\Delta\gamma (T) \,dT \tag{10}$$

$$\int \frac{(c_N - c_s) \,dT}{T} = 0 = \int \Delta\gamma (T) \,dT \tag{11}$$
where $\gamma$ is the quantity $c/T$ plotted in Figure 2, and $c_N$ is the normal and $c_s$ the superconducting specific heat. The total binding energy is considerably smaller than that of YBCO, roughly

$$E_b^0 \simeq 220 \pm 50 \frac{\text{mJ}}{\text{gm atom}}$$

$$= 1.7 \pm 4 \times 10^5 \text{erg/cc}$$

Interestingly, this is below (by a factor of 2) what I would predict from scaling from YBCO by $T_c^2$, perhaps partly because of a contribution from the chains. With less accuracy because of the critical fluctuation effects on $c_s$ for low doping, I can also estimate $E_b$ for the doping levels 13.5% and 10%.

Using the value (13) I obtain $\lambda_c = 3 \pm 1 \mu m$ for optimal doping, which is embarrassingly close. Figure 3 shows my estimates for three doping levels plotted on Uchida’s curve of $\lambda$ as calculated from Figure 1 using Eq. 9, with my estimates plotted as areas that make some attempt to express the uncertainties.

The agreement both as to numerical value and trend is heartening. For 2 1 4, driving a critical Josephson current is precisely sufficient to erase the energy of the superconducting correlation. Undoubtedly it is possible to invent a system of carefully balanced cancellations that would nonetheless ascribe the source of superconductivity to internal correlations in the planes but such logical contortions seem improbable and may even be impossible. Why would an intraplanar mechanism correlate its $T_c$ as well as its energy precisely with the strength of interplanar coupling, over a range of 5 to 1 in $T_c$?

The case of $Hg$ 1 2 0 1 is much less airtight, but still strong. I know of no satisfactory specific heat measurements so we are reduced to scaling the binding energy according to $T_c^2$, and hence $\lambda_c$ according to $T_c$. I predict, then, for $Hg$ 1 2 0 1

$$\lambda_c = 3 \mu m \times \frac{40}{90} \times \frac{d_{214}}{d_{Hg}}$$

$$= 1 \pm 0.5 \mu m$$

The observed value is quoted as $1.34 \mu m \pm$ about 10%. The agreement is spectacular.
$Tl\ 1\ 2\ 0\ 1$ would be predicted, on the same basis, to have $\lambda_c \simeq 0.8\mu m$, since $d$ is even greater than that for $Hg$, but K. Moler et al [5] find that $\lambda_c > 15\mu m$ for the single crystals for which they have imaged vortices, and this figure is in agreement with estimates by van der Marel (and with my own estimates using transport theory). This is clearly a severe anomaly. The above direct evidence for interplanar coupling in the other cases is supplemented by the neutron scattering evidence in YBCO which shows that the gap structure is strongly correlated between planes in the close pair, in just such a way as to optimize interplanar kinetic energy [10]. I cannot emphasize too strongly the need to assure ourselves that $Tl\ 1\ 2\ 0\ 1$ is genuinely a one-layer case. Some evidence for structural defects exists.

The ILT hypothesis for the high $T_c$ cuprates was based from the start on an experimental observation: that $c$-axis conductivity is non-metallic and incoherent where that in the $ab$ plane is metallic, if in many respects very anomalous. This behavior is presumed to be a result of a non-Fermi liquid, charge-spin separated state; but the hypothesis can be directly tested in a manner completely independent of that conjecture. There are two experimentally testable consequences of the idea, if one is able to measure the $c$-axis electrodynamics in the superconducting state, as has been done in a number of cases. The first is violation of the “Josephson identity”, which expresses the fact that in BCS superconductors pair tunneling replaces the coherent normal state conduction. This violation has been noted previously by Timusk. [15] The second is the requirement that the supercurrent kernel $\frac{e}{4\pi^2}\lambda^2$ almost precisely match the condensation energy of the superconductors. It seems to me that this agreement effectively rules out any intralayer theory of high $T_c$, and points to the interlayer concept, for those cases in which it occurs; but we are left at a loss in the one clear case where it does not [16].
FIGURE CAPTIONS

1. Reflectivity measurements from ref. (6)

2. Specific heat of 214 samples. Doping \( x \) is the parameter.

3. The points are the measured values of \( \lambda_c \) from ref. (6); the large ovals are the result of our theory (eq. [6]) including rough estimates of limits of error.
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[16] I should like to acknowledge extensive discussions with D. van der Marel, S. Chakravarty, D.G. Clarke, N.-P. Ong, and especially K.A. Moler.