Where is the energy saved?

A rather generic and unsatisfying feature of all the theoretical scenarios discussed in the last lecture is that, while purporting to explain part or (occasionally) all of the existing theoretical data, they make few if any quantitative predictions about phenomena as yet unobserved. (In a few cases a theory may seem to do so, but on close examination it usually turns out that the 'prediction' is in effect a simple extrapolation of trends already observed and is thus not terribly surprising.) In this lecture I review a couple of theoretical scenarios which, by contrast, have attempted to make quantitative predictions about experimental quantities which at the time of the prediction were not measured, and examine the extent to which the predictions agree with subsequent experiment. The first is the 'inter-layer tunneling' scenario of P.W. Anderson and collaborators, the second my own 'midinfrared' scenario. Both focus primarily on the 'mechanism' of the superconducting transition (in fact, neither as such claims to explain the N state behavior in detail*), and both emphasize, in rather different ways, the question 'where is the energy saved?' One more feature which the two scenarios have in common (and in which they differ from the vast majority of other theoretical proposals in the literature) is that both take very seriously the observed dependence, in homologous series, of $T_c(n)$ on the layer multiplicity n. So it is appropriate to start by saying a word about this:

To recapitulate the results quoted in lecture 6, in the Ca-spaced homologous series (Bi, Tl, Hg) $T_c(n)$ increases with n as far as $n = 3$, thereafter apparently decreasing (at least in the cases, such as the Tl-2 series, where one can be reasonably sure that the material is single-phase). In all the cases quoted, the T_c's for $n \leq 3$ appear to fit rather well the formula

$$\frac{T_c^{(3)} - T_c^{(2)}}{T_c^{(2)} - T_c^{(1)}} = \frac{1}{3} \tag{1}$$

However, we should strongly caution that this apparently impressive coincidence among four different series (Bi, Tl-1, Tl-2, Hg) is almost certainly less significant than it looks, since the ratio α of $T_c^{(2)} - T_c^{(1)}$ to the single-layer transition temperature $T_c^{(1)}$ is quite different for the Bi and Tl-1 series ($\alpha \sim 2$) to that for the Tl-2 and Hg series ($\alpha \sim 0.5$). My own belief is that one should attach significance to the relation (1) only in these cases (Tl-2, Hg, possibly Ca) where $T_c^{(1)}$ is already of order 100K. The above statements refer exclusively to Ca-spaced homologous series: when the spacer is (entirely) Sr or Ba, by contrast, the 2- and 3-layer members are almost invariably nonsuperconducting,† probably though not certainly because of the effect of 'intruder' oxygens.

Clearly, there are two major classes of explanation for the observed behavior of T_c as a function of n.

(1) The mechanism of superconductivity is entirely (or overwhelmingly) confined to the single CuO_2 planes, and the reason that $T_c(n)$ increases with n in the Ca-

*In work which is logically closely related to the ILT scenario, Anderson has claimed to explain the N-state properties. However, no direct quantitative test has been suggested for this part of the scenario.

†But cf. Jim Eckstein's recent result in $Bi_2Sr_3Cu_2O_8$.

spaced series is that the properties of the individual planes are 'improved' in some way in multilayer materials.

(2) The increase of T_c is a result of some kind of interaction between the different CuO_2 planes.

Let's first discuss some possible class-1 explanations. In the early days, it was often postulated that the effect of adding more planes was simply to increase (or otherwise improve) the number of carriers per plane. Certainly, NMR measurements in Hg-2223[‡] do seem to indicate that the number of carriers in the central plane is different (probably less) than that in the outer ones of the trilayer, and this is not entirely unexpected from the point of view of the electrostatics. However, a major argument against attributing the increase in T_c to this effect is simply that it is by now very well established that in all superconducting cuprates, including the single-layer ones like Tl-2201, T_c has a maximum as a function of doping; it then immediately follows that no change in the doping can by itself increase T_c above this maximum. A similar argument refutes the suggestion that adding extra planes changes the buckling; all the evidence is that the 'optimal' buckling is zero, and this is already attained in the one-layer Tl and Hg compounds. A third class-1 proposal is that as a result of adding extra planes the dielectric screening within the individual planes is changed; this is certainly likely to be true, since e.g. for a 3-layer material the rather polarizable oxide layers are replaced, for the middle plane, by very unpolarizable Ca^{++} ions. The hypothesis that it is this effect which is responsible for the increase in T_c is more difficult to refute, and in fact has the advantage that it might go some way towards explaining why the introduction of (highly polarizable) intruder oxygens appears to depress T_c so severely: however, it would not seem to explain why it is depressed way below the one-plane value, nor why in the Ca-spaced compounds T_c decreases for $n \geq 4$. So it seems at least possible that the true explanation of the variation of T_c with n is of class 2; both the principal scenarios to be discussed in this lecture are of this class.

The interlayer tunneling scenario[¶]

The ILT scenario (or, as its proponents would call it, theory) for HTS dates from September 1987, and was vigorously maintained by Anderson and a few collaborators for about ten years thereafter. Although it is by now largely discredited, I believe it is well worth reviewing in some detail, since it seems to me to have many of the generic characteristics that one would hope for in any eventually successful theory, in particular in that it makes very specific and quantitative predictions, and hence is eminently 'falsifiable' in the sense of K.R. Popper.

The ILT scenario rests on two major hypotheses:

[‡]Michalak et al., Physica C **235**, 1673 (1994).

[¶]Ref.: P.W. Anderson, The Theory of Superconductivity in the High-T_c Cuprates, Princeton University Press, 1997, ch. 7.

(1) The behavior of the electrons in a single CuO_2 plane is not describable by Fermi-liquid theory, and as a result hopping of single electrons between neighboring plane is strongly inhibited.

(2) In the superconducting phase, by contrast, Cooper pairs can tunnel relatively freely between neighboring planes, and it is the resulting lowering of the c-axis kinetic energy which is, wholly or dominantly, the source of the superconducting condensation energy.

Regarding hypothesis 1, it is not always very clear exactly what is the correct description of the single-plane state of the electrons: it is variously described as 'RVB', 'spin-charge separated', 'tomographic Luttinger liquid' and other things. What is common to all the variants is that the single-particle in-plane Green's function does not have a simple pole, even in the limit $\omega \to \epsilon_F$ (if indeed the concept of a Fermi energy makes sense!); or in less technical language, that the elementary excitation of the single planes are not FL-type quasiparticles but much more complex entities involving correlation of many electrons. The argument is then that since the inter-plane tunneling term in the Hamiltonian is of the form $\sum_{\mathbf{kk}'} t_{\mathbf{kk}'} a^\dagger_{\mathbf{k}n} a_{\mathbf{k}'n+1}$ + h.c. i.e. it annihilates exactly one electron on plane n and creates one on plane $n+1$ (or vice versa), it is as it were wrongly matched to the in-plane elementary excitation spectrum – to hop successfully, the electron would have as it were to change all the other degrees of freedom on the recipient plane so as to create the necessary many-body excitation. The exact details of the model appear to be somewhat time-dependent, but the generic upshot is that the reduction in total N-state energy due to interplane tunneling, which in a textbook metal would be of order $t_\perp^2 N(0)$ where $t_\perp$ is the order of magnitude of $t_{\mathbf{kk}'}$, is not realized. On the other hand, it is argued, when Cooper pairs form, they can tunnel freely, so the $\Delta\phi$ dependent term in the Josephson energy *is* realized. The crucial point is that, unlike in the standard treatment of the LD model (see lec. 9), there is no constant term, i.e. the saving at (say) $\Delta\phi = 0$ is real.

More formally: let us define a generalized LD model by the statement that to lowest order in the interplane tunneling matrix element $t_\perp$, the Josephson-like energy *relative to the normal ground state* induced by tunneling between neighboring planes with a difference $\Delta\phi$ in the phase of the Cooper-pair wave function is of the form

$$E(\Delta\phi) = K - J\cos\Delta\phi \tag{2}$$

where for the moment K and J are treated as phenomenological constants. In the standard 'Ambegaokar-Baratoff' treatment of the LD model we have $K \sim J$ (with an exact equality in the case of exact particle-hole symmetry), and thus, as already emphasized in lecture 9, interplane tunneling does not lower the energy of the superconducting ground state relative to the normal one. However, it is characteristic (in fact a defining postulate) of the ILT model that K is either zero or $\ll J$, and hence that in the S ground state ($\Delta\phi = 0$) the energy is lowered relative to the normal ground state by an amount J per pair of planes. In general, J may of course be different for inequivalent pairs (e.g. we would expect intuitively that in Bi-2212 J is larger for the pair of planes which

compose a bilayer than for a pair of neighboring unit cells). It is supposed that it is this energy saving which is all, or at least a large part (let us say η) of the superconducting condensation energy $E_{\rm cond}$.

The remarkable thing about this idea is that it is subject to more or less direct experimental test, since in a single-plane material the coefficient J is directly proportional to the (3D) c-axis superfluid density* $\rho_{s\perp}$. In fact, directly from the considerations of lecture 9 we have the relation

$$\Delta T_\perp = \left\{ \frac{\hbar}{2md} \right\}^2 \rho_{s\perp} \tag{3}$$

where $\Delta T_\perp \, (\propto J) \equiv \eta E_{\rm cond}$ is the energy saving per unit volume and d is the interplane spacing. Using the relation between $\rho_{s\perp}$ and the c-axis penetration depth $\lambda_\perp$ and assuming the local magnetic permeability μ to be 1, we obtain a prediction for $\lambda_\perp$ which is conveniently written in the form†

$$\lambda_\perp = \eta^{-1/2} \lambda_{\rm ILT}, \quad \lambda_{\rm ILT} \equiv \left\{ \frac{mc^2}{E_{\rm cond}} \frac{a_0 A}{16\pi d} \right\}^{1/2} \tag{4}$$

where $E_{\rm cond}$ is now the condensation energy per formula unit (i.e. per CuO_2) at $T = 0$, a_0 is the Bohr radius and A the area per formula unit. Note that the formula for $\lambda_{\rm ILT}$ does not involve the poorly known c-axis dielectric constant $\epsilon_\perp$.

It is interesting to compare the prediction (4) with that which would follow from the application to each inter-plane 'junction' of the Ambegaoker-Baratoff formula (part I, lecture 13)

$$I_c R_n = \frac{\pi \Delta}{2e} \tag{5}$$

with Δ taken to have the BCS value $1.76 k_{\rm B} T_c$. If we call the value of $\lambda_\perp$ predicted in this way λ_ρ we have according to the results of lecture 7

$$\lambda_\rho = \left\{ \frac{\hbar c^2 \epsilon_0 R_n}{\pi \Delta} \right\}^{1/2} \frac{1}{2d} \tag{6}$$

We saw in lecture 7 that according to the Basov plot,‡ most of the measured values of $\lambda_\perp$ whether for single- or multilayer cuprates,§ seem to fall close to λ_ρ.

It is clear that if the predicted value of $\lambda_{\rm ILT}$ for a given single-plane cuprate is of the same order as that of λ_ρ, an examination of the experimentally measured $\lambda_\perp$ will not

*The case of a multilayer cuprate is more complicated, since in this case $E_{\rm cond}$ comes predominantly from the 'strongest' tunneling links whereas $\rho_{s\perp}$ is determined primarily by the 'weakest' ones: so there is no one-one correspondence between the two quantities.

†AJL, Science **274**, 587 (1996). Note that I have incorporated, here, an extra factor of 2 into the definition of $\lambda_{\rm ILT}$.

‡D.N. Basov et al., Phys. Rev. B **50**, 3511 (1994).

§In the multilayer case we would expect both $\lambda_\perp$ and $\rho_\perp$ to be diminished by the 'easiest' links, so this result is expected

necessarily be very informative. This is the case for $La_{2-x}Sr_xCuO_4$ at various dopings since both $\lambda_{\rm ILT}$ and λ_ρ lie in the range $3-15\mu$ (cf. Anderson, Science **279**,1197 (1998)). However, the situation is quite different for Tl-2201: in this case λ_ρ is around 20μ, whereas from the measured value of the condensation energy we have $\lambda_{\rm ILT} \sim 0.9\mu$ (note this corresponds to a discrepancy of a factor ~ 500 in the more physically meaningful quantity $\rho_{s\perp}$!). For Hg-1201 the value of λ_ρ is $\sim 10\mu$; the prediction for $\lambda_{\rm ILT}$ is less accurate than for Tl-2201 because the condensation energy is less reliably measured, but $\lambda_{\rm ILT}$ should again be of order 1μ. This is a crucial test of the ILT model, at any rate in the simple form embodied in eqn. (1), should be a measurement of $\lambda_\perp$ for these two materials.* This has been accomplished by Moler, Kirtley and co-workers by a very direct technique (imaging of 'c-axis vortices": note that while the resolution in these experiments is insufficient to measure λ_{ab}, it is more than adequate for the measurement of $\lambda_\perp$). The results are unambiguous:‡‡ $\lambda_\perp \approx 17-21\mu$ for Tl-2201 and ≈ 8 for Hg-1201, in clear agreement with the predicted λ_ρ and order-of-magnitude disagreement with $\lambda_{\rm ILT}$. Thus the ILT scenario, at least in its simplest and most discussed version, seems to be unambiguously refuted.
[Two cautions:

(1) It may be just possible to rescue the scenario if one is prepared to go beyond the simple ansatz (1), but only at the cose of postulating rather wild fluctuations in the c-axis tunneling properties for which there seems to be no obvious independent evidence (see AJL, ref. cit.)

(2) The Moler-Kirtley experiments cannot exclude a much watered-down version of the model, in which most of the condensation energy (in fact all in single-plane materials) comes from (unspecified) in-plane effects but the 'boost' in multilayer materials derives from c-axis tunneling. See S. Chakravarty et al., PRL **82**, 2366 (1999); Phys. Rev. B **67** 100504 (2003)].

In the rest of this lecture I want to discuss a rather different approach to the problem of energy saving.† For simplicity I will start with the case of a single-plane material, and subsequently generalize the discussion to the multiplane case. Consider, then, a single-plane cuprate such as Tl-2201 or Hg-1201. I start from four fundamental assumptions:

(1) The usual separation of the electrons into 'core' and 'conduction' electrons is legitimate, so that we can write down an effective Hamiltonian for the conduction electrons alone, with the effects of the core electrons completely embodied in an external potential and the screening of the inter-conduction electron Coulomb interaction.

*The first indication that $\lambda_\perp$ was $\ll \lambda_{\rm ILT}$ came from the c-axis optical experiments of van der Marel and coworkers, who established the absence of a c-axis plasmon for $\nu > 100{\rm cm}^{-1}$. However, the conversion from ν_ρ to $\lambda_\perp$ involves the c-axis dielectric constant, which has been the subject of some controversy.

‡‡Moler et al., Science **279**, 1193 (1998); PRL **81**, 2140 (1998)

†Ref.: AJL, Proc. Natl. Acad. Sci. **96**, 8365 (1999).

(2) The dominant players in superconductivity are the electrons in the CuO_2 planes.

(3) Ionic motion is irrelevant, at least in the first approximation.

(4) For the purposes of analyzing the mechanism of superconductivity, we may neglect inter-layer tunneling.†

Of these assumptions, (1) and (2) are virtually universal in the literature on the cuprates and probably do not need detailed justification, while (3) may be justified by appealing to the absence of any appreciable isotope effect in the higher-T_c cuprates (lecture 11). Postulate (4) is of course the exact opposite of what is postulated in the ILT model: the simplest justification is to note that if we describe the c-axis transport by an incoherent hopping model with some plane-to-plane hopping rate τ^{-1}, then we can infer τ from the measured value of the c-axis resistivity $\rho_\perp$ (plus a reasonable estimate of the in-plane DOS) and if we do so we find for all single-plane materials $\tau \gg \hbar/k_{\mathrm{B}}T_c$, where the latter is presumably the relevant 'timescale' characterizing the formation of the superconducting state.

It is convenient to make, for the sake of a simple exposition *only*, two further assumptions:

(5) The Coulomb interaction between conduction electrons in different CuO_2 planes may be neglected (in a single-plane material)

(6) The dielectric constant $\epsilon_{\mathrm{core}}(\mathbf{q},\omega)$ which represents the screening of the Coulomb interaction between conduction electrons in the same CuO_2 plane by the 'core' electrons (both in- and off-plane) may be approximated by a real positive constant ϵ_{sc}.

If we accept assumption (6), then a reasonable value of ϵ_{sc} can be inferred from the optical data in the region (say) $2-4$eV, and is of order $4-5$.

In the light of assumption (5), the Hamiltonian is now simply the sum of independent Hamiltonians describing the different CuO_2 planes, and in the light of assumption (6) the Hamiltonian of a single plane may be written

$$\hat{H} = \hat{T}_\parallel + \sum_{\mathbf{k}} \hat{\rho}_{\mathbf{k}} U_{-\mathbf{k}} + \frac{1}{2}\frac{e^2}{2\epsilon_0\epsilon_{\mathrm{sc}}}\sum_{\mathbf{q}} |\mathbf{q}|^{-1}\hat{\rho}_{\mathbf{q}}\hat{\rho}_{-\mathbf{q}} \equiv \hat{T}_\parallel + \hat{U} + \hat{V} \tag{7}$$

where $\hat{\rho}_{\mathbf{q}}$ is the 2D Fourier transform of the conduction-electron density, $\mathbf{k}$ is a reciprocal lattice vector and $\hat{T}_\parallel$ is the (true) in-plane kinetic energy. This Hamiltonian is very generic, and most of the models used as starting points in the literature (Hubbard, $t-J$ ...) can be regarded as simplifications or special cases of it. Note that it is *complete*: if one believes that e.g. spin fluctuations, anyons or excitons play an important role, they have to be *derived* from (7), not added to it!

Imagine that we could somehow cool the system described by (7) down to zero temperature but forbid it, by fiat, to form Cooper pairs (or undergo any other phase

†Except in so far as it will eventually be necessary to stabilize genuine 3D long-range order.

transition), i.e. we assume it to find the best 'normal' ground state.[¶] If we now relax the constraint, we know from experiment that the system will form pairs, and the reason it does so, at $T = 0$, is, trivially, to reduce its total energy. It follows that at least one of $\hat{T}_{\|}$, $\hat{U}$ and $\hat{V}$ must decrease, possibly though not necessarily at the expense of an increase in the other two. Let us suppose, for the sake of definitions, that $\langle \hat{V} \rangle$ decreases (without any assumption about $\langle \hat{T}_{\|} \rangle$ and $\langle \hat{U} \rangle$; nothing actually depends on this. Now, as we shall see in a moment, the value of $\langle \hat{V} \rangle$ in an arbitrary state of the system can be expressed as a sum rule, i.e. as a sum of contributions from different regions of wave vector $\mathbf{q}$ and frequency ω: so a natural question is: *In what regime(s) of* $\mathbf{q}$ *and* ω *does the saving*[**] *of Coulomb energy occur?* Oddly enough, until recently this question seems hardly to have been asked in the literature.

Let's try to be a bit more quantitative. An exact expression for the Coulomb energy is

$$\langle \hat{V} \rangle = \frac{1}{2} \sum_{\mathbf{q}} V_{\mathbf{q}} \langle \hat{\rho}_{\mathbf{q}} \hat{\rho}_{-\mathbf{q}} \rangle = \frac{1}{2\pi} \sum_{\mathbf{q}} \int_0^\infty d\omega \, \mathrm{Im}\, \chi(\mathbf{q}, \omega), \qquad (V_{\mathbf{q}} \equiv e^2 / 2\epsilon_0 \epsilon_{\mathrm{sc}} |\mathbf{q}|) \tag{8}$$

where $\chi(\mathbf{q}, \omega)$ is the complete (true) density-density correlation function of the system. Note that this expression is exact, independently of the effects of lattice structure. However, the latter now leads to a complication: we would like to express $\chi(\mathbf{q}, \omega)$ in terms of $V_{\mathbf{q}}$ and the 'bare' correlation function $\chi_0(\mathbf{q}, \omega)$, by which we mean the quantity defined diagrammatically by omitting all those graphs in $\chi(\mathbf{q}, \omega)$ which can be cut into two by cutting a single Coulomb line of momentum $\mathbf{q}$. The problem is that χ_0 is actually a *matrix* in the reinforced space, i.e. it is specified by two arguments, $\mathbf{q}$ and $\mathbf{q} + \mathbf{K}$, and this leads to rather a messy form for $\hat{V}$ (see appendix B of AJL, ref. cit.). For pedagogic simplicity I will therefore assume at this point that the matrix can be approximated by its diagonal terms: while in the general case this approximation may introduce some error, it can be shown (ref. cit.) that it does not affect appreciably the arguments I shall give concerning the long-wavelength limit.

With the above approximation we then have

$$\chi(\mathbf{q}, \omega) = \frac{\chi_0(\mathbf{q}, \omega)}{1 + V_{\mathbf{q}} \chi_0(\mathbf{q}, \omega)} \tag{9}$$

and inserting this into the expression for $\langle \hat{V} \rangle$

$$\langle \hat{V} \rangle = -\frac{1}{2\pi} \sum_{\mathbf{q}} \int d\omega \, \mathrm{Im} \left(1 + V_{\mathbf{q}} \chi_0(\mathbf{q}, \omega) \right)^{-1} \tag{10}$$

Note that formula (10) is actually valid in any number of dimensions, provided that $V_{\mathbf{q}}$ has the appropriate form. In particular, in the bulk 3D case we have $V_{\mathbf{q}} = e^2 / \epsilon_0 \epsilon_{\mathrm{sc}} \mathbf{q}^2$,

[¶] The nearest approximation to this in real life is to cool in a strong magnetic field, as in the experiments of Boebinger.

[**] Or increase, if $\langle \hat{V} \rangle$ indeed increases.

and since the (longitudinal) dielectric constant $\epsilon_{\|}(\mathbf{q},\omega)$ is conventionally defined as* $1+(e^2/\epsilon_0\epsilon_{\rm sc}\mathbf{q}^2)\chi_0(\mathbf{q},\omega)$, eqn. (10) takes the simple form

$$\langle\hat{V}\rangle = -\frac{1}{2\pi}\sum_{\mathbf{q}}\int d\omega\,\mathrm{Im}\,\frac{1}{\epsilon_{\|}(\mathbf{q},\omega)} \tag{11}$$

where the integrand $-\,\mathrm{Im}\,\frac{1}{\epsilon_{\|}(\mathbf{q},\omega)}$ is usually called the loss function and is directly measured in transmission EELS experiments. In 2D one has to be a little more careful, since no true analogy to $\epsilon_{\|}$ exists: it is most convenient to write expression (10) in the form

$$\langle\hat{V}\rangle = -\frac{1}{2\pi}\sum_{\mathbf{q}}\int_0^\infty d\omega\,\mathrm{Im}\,(1+qK(\mathbf{q},\omega)/\epsilon_{\rm sc})^{-1} \tag{12}$$

where the quantity $K(\mathbf{q},\omega)$, which has the dimensions of length, is related to the *3D* dielectric constant by

$$K(\mathbf{q},\omega) = \frac{1}{2\pi}\bar{d}(\epsilon_{\|}(\mathbf{q},\omega)-\epsilon_{\rm b}) \tag{13}$$

where $\bar{d}$ is the interplane separation and $\epsilon_{\rm b}$ is the 'background' (non-conduction-electron) contribution† to $\epsilon_{\|}(\mathbf{q},\omega)$.

Let's now consider the contribution to the fundamental expression (10) for $\langle\hat{V}\rangle$ from different regions of $\mathbf{q}$ (in the N phase). To do this, it is convenient to note the Kramers-Kronig relation for $\chi_0(\mathbf{q},\omega)$:

$$\frac{1}{\pi}\int\frac{\mathrm{Im}\,\chi_0(\mathbf{q},\omega)}{\omega}\,d\omega = \chi_0(\mathbf{q}) \tag{14}$$

where $\chi_0(\mathbf{q})$ is the 'bare' static susceptibility, which we expect to be not too strongly varying with $\mathbf{q}$ and of the order of magnitude of $\chi_0(0)\sim dn/d\epsilon$. From this relation we see that in general‡ is we would expect $\mathrm{Im}\,\chi_0(\mathbf{q},\omega)$ to be of the general order of magnitude of $dn/d\epsilon$ (and then, by the KK relation applied to general ω,the real part should be of the same order of magnitude). This then leads us to define a 2D 'Thomas-Fermi' wave vector $q_{\rm TF}$ by the relation

$$V_{q_{\rm TF}}\left(\frac{dn}{d\epsilon}\right) = 1 \tag{15}$$

For a noninteracting-band model with effective mass m^*, we have $dn/d\epsilon = m^*/\hbar^2$ and so

$$q_{\rm TF} = 2(m^*/m)\,\epsilon_{\rm sc}^{-1}a_0^{-1} \tag{16}$$

(a_0 = Bohr radius). With $m^*/m\sim\epsilon_{\rm sc}\sim 4$ this gives $q_{\rm TF}\sim 4\text{Å}^{-1}$, *irrespective* of the density of carriers. Note incidentally that in 3D we can apply the same general arguments, and obtain the standard result

$$q_{\rm TF} = \left[\frac{e^2}{\epsilon_0\epsilon_{\rm sc}}\left(\frac{dn}{d\epsilon}\right)\right]^{1/2} = \left[\frac{2}{\pi}\,\frac{m^*}{m}\,\frac{1}{\epsilon_{\rm sc}a_0}\right]^{1/2}k_{\rm F}{}^{1/2} \tag{17}$$

*Modulo some rather messy questions concerning the treatment of the factor $\epsilon_{\rm sc}$.

†The equality $\epsilon_{\rm b}=\epsilon_{\rm sc}$ holds only in specific models.

‡These arguments need some modification in the limit $\mathbf{q}\to 0$, cf. below.

The above estimates should be understood in an order-of-magnitude sense only, but what is interesting is that in both the 2D and 3D cases $q_{\rm TF}$ is of the general order of magnitude of the Fermi wave vector $q_{\rm F}$ (and certainly not much smaller).

The significance of the quantity $q_{\rm TF}$ is that it separates two regimes where the effect of changing $\chi_0(\mathbf{q},\omega)$ may be qualitatively different. Consider first the regime $q \gg q_{\rm TF}$. In this regime, by the above arguments, we have $|V_{\mathbf{q}}\chi_0(\mathbf{q},\omega)| \ll 1$, and thus can expand the integrand in the expression for $\langle\hat{V}\rangle$:

$$\langle\hat{V}\rangle_{q\gg q_{\rm TF}} \approx +\frac{1}{2\pi}\int d\omega\, V_{\mathbf{q}}{\rm Im}\,\chi_0(\mathbf{q},\omega) = V_{\mathbf{q}}\langle\hat{\rho}_{\mathbf{q}}\hat{\rho}_{-\mathbf{q}}\rangle_0 \tag{18}$$

where the quantity $\langle\hat{\rho}_{\mathbf{q}}\hat{\rho}_{-\mathbf{q}}\rangle_0$, which is essentially defined by the $(\ldots)$-equality, is the (hypothetical) 'bare' static correlation. This is of course exactly what one would get from straightforward perturbation theory, and one sees that in this regime the only way to decrease the contribution to $\langle\hat{V}\rangle$ is to *decrease* the quantity $\langle\hat{\rho}_{\mathbf{q}}\hat{\rho}_{-\mathbf{q}}\rangle_0$. In any theory which is remotely of BCS-like type, formation of Cooper pairs with an order parameter which is uniform in sign can only *increase* the 'bare' static correlation since

$$\langle\hat{\rho}_{\mathbf{q}}\hat{\rho}_{-\mathbf{q}}\rangle \sim \sum_{\mathbf{k}} F^*_{\mathbf{k}+\mathbf{q}/2}F_{\mathbf{k}-\mathbf{q}/2} \tag{19}$$

and one thus reaches the conclusion that if indeed Coulomb energy is to be saved in this regime, the quantity $F_{\mathbf{k}}$ *must* (plausibly) *change sign, as a function of* $\mathbf{k}$. Thus it is not particularly surprising that a state of the $d_{x^2-y^2}$ type is found (although of course it is not uniquely selected by this argument).

The opposite, 'overscreened' regime, $q \ll q_{\rm TF}$ (so that $|V_{\mathbf{q}}\chi_0(\mathbf{q},\omega)| \gg 1$) is more complicated to analyze, since in general the contribution to $\langle\hat{V}\rangle$ from this regime depends on both the real and imaginary parts of χ_0. However, one can see that a *uniform* increase in χ_0 will lead to a *decrease* in $\langle\hat{V}\rangle$; physically, this reflects the fact that the screening of the Coulomb interaction becomes more effective and the energy is thereby lowered.

A very important point, which I believe is not widely appreciated, is that the small-q ('overscreened') regime is much more important relative to the phase space as a whole in 2D than in 3D. In 3D, the phase space goes as $q^2\,dq$, and there is no particular compensating factor in the loss function $-{\rm Im}\left(\epsilon_{\|}(\mathbf{q},\omega)\right)^{-1}$; in 2D, by contrast, not only is the phase-space factor only $q\,dq$, but the q is cancelled by the behavior of the Coulomb matrix element in the denominator. Thus, to a first approximation, *all values of q contribute equally.*

My personal conjecture is that the main regime in which Coulomb energy is saved by the formation of Cooper pairs is that of small q (specifically, say, $q \lesssim 0.5\text{Å}^{-1}$). If we grant that, then the next question is: Where is ω? To answer this we use the 'Willie Sutton principle": you can't take it away if it isn't there in the first place! And in the N phase, if we confine ourselves for the moment to $\omega \lesssim 3$eV, the place where the Coulomb energy is predominantly, as seen in the experimentally measured loss function, is the MIR peak, $0.1\text{eV} \lesssim 1-1.5\text{eV}$. Thus the conjecture is that the saving is predominantly from *small q and midinfrared ω* ('MIR scenario').

An advantage of the 'small-q' assumption is that it permits us to explain the behavior of $T_c(n)$ in multilayer cuprates in terms of the effects of the inter-plane Coulomb interaction (which must be there!). See AJL, PRL **83**, 392 (1999), where it is shown inter alia that this hypothesis leads to the formula, valid for 'not too large' n,

$$T_c(n) - T_c(1) = \text{const } (1 - 1/n) \tag{20}$$

The ideal test of the MIR scenario would he differential transmission EELS experiments, but these turn out to be technically very difficult.

Thus, we do not have direct measurements of the change in the bare density response function χ_o(q,ω) (hence of the Coulomb energy) through the superconducting transition. On the other hand, there have been a number of experiments on the change of the optical properties through the transition. What the optical experiments measure, in principle, is the complex <u>transverse</u> dielectric constant $\epsilon_\perp(\mathbf{q},\omega)$, whereas the bare density response function is simply related (by $\epsilon_\parallel(\mathbf{q},\omega) \equiv 1 + (e^2/\epsilon_o q^2)\chi_o(q\omega)$) to the <u>longitudinal</u> dielectric constant $\epsilon_\parallel$(qω). Now in the limit $q \to 0$, and in the absence of off-diagonal long-range order, there is a theorem that the quantities $\epsilon_\parallel$(qω) and $\epsilon_\perp$(qω) must coincide for any ω (basically in this limit the system "cannot tell" whether the local electromagnetic current is longitudinal or transverse). In the presence of ODLRO the equality no longer holds in general (cf. the difference between $\epsilon_\parallel$(q0) and $\epsilon_\perp$(q0) which underlies the Meissner effect), but since the difference can be traced to the Anderson-Bogoliubov mode of the (hypothetical) neutral condensate, one may hope that in the limit $\omega/\mathrm{v}_F\mathrm{q} \gg 1$ it vanishes to a good approximation. If this is so, then by inspection of the optical data one should be able to infer the changes in χ_o(qω) and thus in the part of the Coulomb energy which is locked up at the (small relative to k_F) values of q involved in the optics.

The earlier optical data* showed that when the system goes through the superconductivity transition, a certain amount of the spectral weight (that is, of the optical conductivity $\sigma(\omega) \equiv \omega\ \mathrm{Im}\epsilon_\perp(\omega)$) is transformed from the frequency region >10,000 cm^{-1}($\sim$ 1 eV) into the low-frequency region ($\omega \lesssim 600$ cm^{-1}). Originally, it was thought that the transfer was from the band 10,000-25,000 cm^{-1}. However, the more recent and accurate experiments of Kuzmenko et al.† measured the real and imaginary parts of the dielectric constant $\epsilon_\perp$(qω) directly by ellipsometry between 0.6 and 2.4 eV, and then used KK relations to infer it over the remaining regimes.

The results are remarkable (see fig. 3 of their paper): over the whole region 0.6-2.5 eV the imaginary part of $\epsilon(\omega)$ (i.e. the optical conductivity) undergoes no change within the error bars at the N-S transition. However, the <u>real part</u> $\epsilon_1(q\omega)$ suffers a noticeable decrease, $\delta\epsilon_1(q\omega) < 0$ (i.e., since ϵ_1 is negative, at least for the lower part of this regime, its absolute magnitude is increased); the greatest decrease occurs at the lower end ($0.6 < \omega < 1eV$) This result is very striking, for the following reason: As we have seen, over the whole of the MIR regime, the phase of $\epsilon(q\omega)$ is very close to $3\pi/4$. If one now assumes that the change in ϵ at the N $\to$ S transition is small compared to the

*H. Molegraaf et.al., Science (2002), and earlier references cited therein.

†Phys. Rev. B**72**, 144503 (2005)

original N-state value, we can ask: What is the optimal way to decrease $-\mathrm{Im}(1/\epsilon)$? And the answer is: Keep $\mathrm{Im}\epsilon$ constant, but decrease $\mathrm{Re}\epsilon$! So the data is at least qualitatively consistent with the idea that an appreciable part of the energy saving takes place in the MIR regime. Whether this saving is large enough to account for the condensation energy, and what exactly is the role of the higher-(frequency (say 1 eV $< \omega <$ 2.5 eV) regime, is at the time of writing unclear.