Cuprate Superconductivity: Dependence of T_c on the c-Axis Layering Structure

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Without relying on any "model" of the in-plane behavior of the electrons in either the normal or the superconducting state, I show that for identically doped members of the same homologous series, as a result of the interplane Coulomb interaction, the difference $\Delta T_c^{(n)}$ of the transition temperature for the nth member from the single-layer value is given by $\Delta T_c^{(n)} = \text{const} \times (1 - 1/n)$. On taking the constant from experiment, I predict large changes in the electron-energy-loss spectroscopy and optical properties in the midinfrared regime on entering the superconducting state, and further infer that the basic mechanism of superconductivity in the cuprates has a large contribution from the small-q regime.

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One of the most intriguing aspects of superconductivity in the cuprates is the apparently systematic dependence of the transition temperature T_c on the c-axis structure and, in particular, on the number n of closely spaced CuO_2 planes. If we consider the homologous series at a given constant level of doping, then the situation may be summarized as follows: In cases such as the Bi, Hg, and various Tl series, when the intra-multi-layer spacer is Ca, T_c rises with n at least as far as $n = 3$ and sometimes up to $n = 5$, thereafter decreasing slowly. On the other hand, when the intra-multi-layer spacer is other than Ca, and, in particular, when it is Sr or Ba, the limited evidence available is compatible with the hypothesis that T_c is actually reduced below the single-plane value and may even be suppressed to zero. A possible reason for this striking difference is proposed in Ref. [1].

In this Letter I restrict myself to the Ca-spaced materials and show that if one assumes no intra-multi-layer tunneling on the relevant time scale [1] it is possible, on the basis of very general considerations, to make a striking *quantitative* prediction about the dependence of T_c on n, and further, on the basis of the comparison of this prediction with experiment, to make a semiquantitative prediction concerning the optical and electron-energy-loss spectroscopy (EELS) properties of these materials in the superconducting state. It should be stressed that these predictions *are completely independent of any "model"* either of the normal state of the electrons in the individual CuO_2 planes or of the fundamental mechanism of superconductivity in the cuprates; nevertheless, to the extent that they are confirmed, they permit an important conclusion concerning the latter. Ideally this Letter should be read in conjunction with Ref. [1].

The basic physical idea is that the total Coulomb energy in an n-layer structure may be thought of as locked up in different modes of charge oscillation which, while far too broad in spectrum to qualify as "plasmons," can nevertheless be classified by their symmetry; e.g., for $n = 2$ one has one "optical" (in-phase) mode and one "acoustic" one, and in general $n - 1$ acoustic modes. If we focus on wavelengths of the order of the inverse interplane spacing and longer, all oscillations are strongly overscreened, so the Coulomb energy associated with a given mode is, roughly speaking, inversely proportional to the strength of screening; this quantity in turn is the product of the "bare" density response (essentially the K_0 introduced below), which within our ansatz is a one-plane quantity and thus independent of n, and an effective coupling constant [$\propto$ the $f_i(q)$ below] which for $q \to 0$ is smaller for the acoustic modes than for the optical ones, so that in this limit the former dominates the energetics. Now, the effect of Cooper pairing is to modify (in general to increase) the bare response, which increases the efficiency of screening and thus *decreases* the Coulomb energy, thereby providing the energy saving which drives the superconducting transition. It turns out (not obviously) that the saving of Coulomb energy in an n-layer structure, which has to be shared between the n planes, is simply proportional to the number of acoustic modes; thus the saving per layer is proportional to $1 - 1/n$. To the extent that the fractional change in T_c with n is small, it should be proportional to the saving, and thus we obtain Eq. (5) below. A more detailed physical discussion is given in Ref. [2].

In the following I denoted by d, $d_{\rm int}$, and $\overline{d}$, respectively, the intra-multi-layer spacing ($\sim$3.5 Å), the inter-multi-layer (center-to-center) distance (typically 6–15 Å), and the average interlayer spacing. Further, I introduce a quantity q_0 which is an order-of-magnitude measure of the typical scale of the midinfrared (MIR, cf. below) normal-state in-plane behavior. Depending on what we believe to be an appropriate model for this behavior, it might be reasonable to take q_0 as, e.g., the primitive reciprocal lattice vector, the Fermi wave vector (if it exists), or something else, but the important point for our purposes is that unless the normal state is very anomalous, q_0 is unlikely [3] to be less than about 1 Å^{-1}. Thus the quantity $q_0 d_{\rm int}$ is always much larger than 1, and the quantity $q_0 d$ is at least 3–4, a number I shall treat for present purposes as "large."

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The analysis of this Letter rests on three fundamental assumptions:

(A1) The explanation of high-temperature superconductivity in the cuprates is to be sought in the interactions of the electrons in the CuO_2 planes with one another and with the static lattice.

(A2) Once the number of carriers per CuO_2 plane (and the structure of the plane) is specified, the bare [4] normal-state in-plane properties (or at least those relevant to superconductivity) are universal.

(A3) The c-axis transport of charge both between and (in Ca-spaced materials) within multilayers is too slow to be relevant to superconductivity.

Assumption (A2) is in principle testable by experiment, and despite uncertainties connected with the contribution of the off-plane "background," it is probably fair to say it is at least consistent with existing data. For (A3), cf. Ref. [1].

I shall now, for brevity of presentation *only*, make four additional assumptions; for reasons of space I shall simply state here that relaxing these does not change the outcome qualitatively, and that in most cases even the quantitative change resulting is small [2].

(A4) The quantity $(q_0 d_{\text{int}})^{-1}$ is sufficiently small that it may safely be set equal to zero.

(A5) Both (i) the effect of interplane Coulomb interactions relative to intraplane ones and (ii) the relative change of (the important elements of) in-plane response functions between the normal and superconducting states are sufficiently small that we may work to lowest nonvanishing order in each.

(A6) The bare normal-state in-plane density response function [4] $\chi_0(\mathbf{r}, \mathbf{r}', \omega)$ may for the purposes below (*only*) be taken to be a function only of the difference $\mathbf{r} - \mathbf{r}'$, so that its Fourier transform $\chi_0(\mathbf{q}, \omega)$ may be labeled by a single variable $\mathbf{q}$ (rather than being a matrix with respect to the reciprocal lattice).

(A7) The effective Coulomb interaction between electrons, whether in the same or different CuO_2 planes, may be approximated by the expression $V_{\text{eff}}(\mathbf{r}) = e^2/(4\pi\varepsilon_0\varepsilon_\infty|\mathbf{r}|)$, where ε_∞ is an appropriate "high-frequency" dielectric constant which takes into account phenomenologically the screening effect of all the core electrons, whether inside, between, or outside the planes.

Using (A1), (A3), and (A4), we see that different multilayers are effectively noninteracting, and moreover that the quantity χ_0 is diagonal in the layer index i; thus the effective Hamiltonian for a single multilayer reads $\hat{H} = \hat{T}_\parallel + \hat{U} + \hat{V}_c$, where $\hat{T}_\parallel$, $\hat{U}$, and $\hat{V}_c$ denote, respectively, the in-plane kinetic energy, the potential energy of the conduction electrons in the field of the in-plane atomic cores, and the interconduction electron Coulomb interaction as screened by the cores [both in and off plane, cf. (A7)]. The observation which is crucial to the argument below is that $\hat{T}_\parallel$ and $\hat{U}$ are simple sums of "single-plane" terms, which by (A2) are universal (and, in particular, independent of the layer multiplicity n), and hence so is χ_0, whereas $\hat{V}_c$ contains both intraplane and interplane terms. In fact, the Fourier transform with respect to the ab-plane component $\mathbf{r} - \mathbf{r}'$ of $V_c(|\mathbf{r} - \mathbf{r}'|)(\mathbf{r}\varepsilon i, \mathbf{r}'\varepsilon j)$ between electrons in planes i and j may be expressed in the form of a matrix,

$$V_{ij}(q) = \frac{\varepsilon^2}{2\varepsilon_0\varepsilon_\infty q} \exp -qd|i - j|, \qquad (1)$$

where $\mathbf{q}$ is a 2D vector in the ab plane ($q \equiv |\mathbf{q}|$). We see immediately that, given the inequality $q_0 d \gg 1$, all effects of the layer multiplicity come entirely from the region of "small" $q (q \ll q_0)$.

I now come to the crux of the argument. I consider a specific homologous series and *assume* that the conditions of comparison are such that the number of carriers per plane is the same in the different members of the series: call this assumption (A8). Given assumptions [A5(i)], (A6), and (A8), the expectation value $\langle V_c \rangle_m$ of the Coulomb interaction for a given multilayer at $T = 0$ may be written quite generally in the form

$$\langle V_c \rangle_m = -\sum_{i=1}^{n} \sum_{\mathbf{q}} \int_0^\infty \frac{d\omega}{2\pi} \times \operatorname{Im}[1 + q f_i(q) K(\mathbf{q}, \omega)/\varepsilon_\infty]^{-1}, \qquad (2)$$

where $f_i(q)$ $(i = 1, 2, \ldots, n)$ is the ith eigenvalue of the matrix $M_{ij}(q) \equiv \exp -qd|i - j|$, and $K(\mathbf{q}, \omega)$ is related to the bare density correlation function [4] $\chi_0(\mathbf{q}, \omega)$ by the formula $K(\mathbf{q}, \omega) \equiv (e^2/2\varepsilon_0 q^2)\chi_0(\mathbf{q}, \omega)$. The single-plane case is, of course, a special case of (2) with $f_1(q) \equiv 1$. From its definition and assumption (A2), $K(\mathbf{q}, \omega)$ is a universal quantity; however, the simplest way of obtaining it experimentally is via a nonuniversal quantity, the measured bulk ab-plane longitudinal dielectric constant $\varepsilon_\parallel(\mathbf{q}, \omega)$, to which it is related by the formula $K(\mathbf{q}, \omega) = \frac{1}{2}\bar{d}[\varepsilon_\parallel(\mathbf{q}, \omega) - \varepsilon_b]$ where ε_b is the contribution to the measured dielectric constant of everything except the conduction electrons in the CuO_2 planes (note that ε_b need not necessarily be equal to ε_∞). Alternatively, $K(\mathbf{q}, \omega)$ may be obtained more directly from EELS experiments: for a single-plane material transmission EELS with $q_z = 0$ and $q d_{\text{int}} \gg 1$ (and the Rutherford factor q^{-4} extracted as is conventional) simply measures q times the integrand of (2), while for a multilayer compound one should also be able to extract K, although the relation is more complicated [5]. As emphasized in Ref. [1], both optical and EELS evidence shows that for small q the quantity $-\operatorname{Im} K^{-1}$ is large only in the midinfrared region (roughly 0.1–1.5 eV) and above; at low energies it is very small.

I next consider the *change* $\delta\langle V_c \rangle^{(n)}$ in V_c for a multilayer system when the system passes (notionally) from the normal to the superconducting state at $T = 0$. Expressing it in terms of an energy per unit area per plane and subtracting the value for the single-plane system, and defining for convenience a (complex) quantity $\eta(\mathbf{q},\omega) \equiv \delta K(\mathbf{q},\omega)/K_0(\mathbf{q},\omega)$ where K_0 is the normal-state value, we obtain using (A5) the expression

$$\delta\langle V_c\rangle^{(n)} - \delta\langle V_c\rangle^{(1)} = \frac{1}{4\pi^2\varepsilon_\infty}\int\frac{d\theta}{2\pi}\int_0^\infty d\omega\int_0^\infty q^2dq\,\mathrm{Im}\left\{n^{-1}\sum_{i=1}^{n}\left[\frac{f_i(q)\eta(\mathbf{q}\omega)K_0(\mathbf{q}\omega)}{[1+qf_i(q)K_0(\mathbf{q}\omega)/\varepsilon_\infty]^2}\right]\right\} - [f_i(q)\to 1]. \quad (3)$$

Equation (3) is exact within assumptions (A1–A8). Note that because of assumption (A5), not only $K_0(\mathbf{q},\omega)$, but also $\eta(\mathbf{q},\omega)$ is universal; thus the effects of the interlayer interactions enter only through the eigenvalues $f_i(q)$.

To proceed further, we need to know something about the magnitude and q dependence of the functions $K_0(\mathbf{q},\omega)$ and, to a lesser extent, $\eta(\mathbf{q},\omega)$. While uncertainties about the quantity ε_b, etc., and the lack of absolute calibration of existing EELS measurements [6] in the relevant (midinfrared) region make it difficult to infer the exact form of K_0 rigorously, all the evidence seems compatible with the hypothesis (which is rather plausible on *a priori* grounds) that this function has little q dependence [2] for $qd \lesssim 1$, and moreover (and this is crucial) that for almost if not all relevant ω we have $|K_0(\mathbf{q},\omega)| \gg d$. Further, while without a detailed theory of the pairing process we evidently have no *a priori* knowledge of the specific form of $\eta(\mathbf{q},\omega)$, strong general arguments [2] suggest that, as in the simple BCS case, it should be constant as a function of q in the limit $q\to 0$, and I shall assume that this behavior persists up to $qd \sim 1$. Given the above information, we see that the dominant contribution to the integral over $\mathbf{q}$ in (3) will come from the region $|K_0(\mathbf{q},\omega)|^{-1} \ll q \ll d^{-1}$. Since in the approximation of tetragonal symmetry K_0 and η must be isotropic in the plane in the limit $q\to 0$, we may write them simply as functions of ω in this region and thus obtain from (3) the simpler expression,

$$\delta\langle V_c\rangle^{(n)} - \delta\langle V_c\rangle^{(1)} = \frac{\varepsilon_\infty}{4\pi^2}\int_0^\infty d\omega\,\mathrm{Im}\left[\frac{\eta(\omega)}{K_0(\omega)}\right] \times\int_0^\infty dq\left\{n^{-1}\sum_{i=1}^{n}f_i^{-1}(q) - 1\right\}. \quad (4)$$

Now, the quantity in brackets in (4) turns out to be given, for all n, by the expression [7] $2(1-1/n)[\exp(2qd)-1]^{-1}$, and the logarithmic divergence of the integral at small q [which must, of course, be cut off by returning to the exact form (3)] should introduce, at least for small n, at best a weak n-dependence. Moreover, in view of assumptions (A2) and [A5(ii)], the increase of T_c in the n-layer system relative to the single-plane one should be simply proportional to minus the (negative, cf. below) left-hand side of Eq. (4). Consequently we obtain, for the relevant class of the homologous series, the strikingly simple prediction ($\Delta T_c^{(n)} \equiv T_c^{(n)} - T_c^{(1)}$),

$$\Delta T_c^{(n)} = \text{const}\times(1-1/n). \quad (5)$$

In attempting to compare the prediction (5) with existing experimental data, the main problem is to identify those cases, if any, in which assumption (A8) is satisfied. Since a plausible, if not certain, indicator of the number of carriers per plane is $R \equiv \lambda_{ab}^{-2}\overline{d}$ (λ_{ab} = in-plane London penetration depth), one would ideally like to compare members of the same homologous series at equal values of R; however, the available measurements of λ_{ab} are insufficient for a systematic comparison. Since there is no space to discuss this question further, I shall err on the side of extreme conservatism and make the comparison only for the $n = 1,2,3$ members of the Hg and Tl $(2,2,n-1,n)$ series, at optimal doping, cases in which I believe the existing data are, at least, compatible with the hypothesis of constant R. Then the prediction of (5) is that $(T_c^{(3)} - T_c^{(2)})/(T_c^{(2)} - T_c^{(1)}) = \frac{1}{3}$ for both series. The experimental ratios are 0.25–0.28 for the Hg series and 0.25–0.34 for Tl. Given the approximations [see especially (A5)] used in obtaining the theoretical prediction, this would seem to indicate that the idea of attributing the systematics of T_c in the Ca-spaced cuprates wholly or mainly to the interlayer Coulomb coupling is, at least, not obviously unviable.

If the observed rise in T_c with n is indeed due to interlayer Coulomb interactions, then Eq. (4) permits a further quantitative prediction: Assuming that the differential loss in Coulomb energy between the N and S states is not actually *less* than the differential increase per unit area per plane in condensation energy, $\Delta E_{\mathrm{cond}}^{(n)}$ (certainly true in BCS theory, and more generally overwhelmingly plausible) we find [8], putting $n = 2$ in Eq. (4),

$$-\delta\int_{\mathrm{MIR}} d\omega\,\mathrm{Im}\left[-\frac{1}{\varepsilon_{\parallel}(\omega)-\varepsilon_b}\right] \geq \frac{8\pi^2 d\overline{d}(\Delta E_{\mathrm{cond}}^{(2)} - \Delta E_{\mathrm{cond}}^{(1)})}{\hbar\varepsilon_\infty\ln(\overline{K}/2d\varepsilon_\infty)}, \quad (6)$$

where both sides of the equation refer to the bilayer compound, δ indicates the change in going from the normal to the superconducting state, and $\overline{K}$ is an appropriate average of $K(\omega)$ over the MIR region. For Tl-2212 we find, taking the logarithm to be of the order of 2 and $\varepsilon_\infty \sim 4$, a value of the right-hand side of (6) of the order of 3 meV/$\hbar$. While optical experiments measure Im$\varepsilon_{\parallel}^{-1}$ rather than the left-hand side of (6), and that somewhat indirectly, it should in principle be possible to verify this inequality directly in EELS experiments [2]; note that the fractional change predicted is at least 2 orders of magnitude greater than the naive estimate $(\sim kT_c/\hbar\omega)^2$ based on simple BCS theory. The inequality (6) rests on the approximation of replacing (3) by (4): A more rigorous inequality can obviously be obtained directly from (3).

Finally, I note that the considerations of this Letter, if correct, have important implications for the basic mechanism of high-temperature superconductivity. Let us consider an optimally doped single-plane cuprate such as Tl-2201 and ask: Suppose that we could somehow exclude all contributions to the saving $\delta\langle V_c\rangle^{(1)}$ of the Coulomb energy [the "1" term in Eq. (6)] from small q, say $q \lesssim d^{-1}$, where d is the Tl-2212 value. How much would T_c decrease? From Eq. (4) and assumption (A5) we see that the answer is approximately $[2/\ln(\overline{K}/2d\varepsilon_\infty)]\Delta T_c^{(2)}$; while we have no good *a priori* estimate of $\overline{K}$, the logarithm is unlikely to be much greater than 2 (and may indeed be less), and $\Delta T_c^{(2)}$ for the Tl-22, $n-1$, n series is about 25 K, so the answer is around 25 K, i.e., about 25% of the original value. Thus even values of $q \lesssim d^{-1}$ make a substantial contribution to the in-plane mechanism, and indeed the above analysis is quite compatible with the original hypothesis of Ref. [1], namely, that the bulk of the mechanism is associated with values of q small compared to q_0, and with values of ω in the MIR region [9].

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[1] A. J. Leggett, J. Phys. Chem. Solids **59**, 1729 (1998). Note, however, that to the extent that the remarks following Eq. (7) of this reference concerning the q dependence of $\delta\chi(q,\omega)$ are inconsistent with those below about $\eta(q,\omega)$, I believe the latter are to be preferred. As a result, the reduction of the effective coupling g due to intra-multi-layer conduction is probably somewhat overestimated in this reference. Note also that in it ε_∞ and ε_b are implicitly identified.

[2] A. J. Leggett, Proc. Natl. Acad. Sci. U.S.A. (to be published).

[3] I believe that "stripes," even if present, are likely to be irrelevant to the MIR behavior.

[4] Defined by omitting from the complete set of diagrams for $\chi(q,\omega)$ all graphs which can be cut into two by cutting a single Coulomb line of wave vector q.

[5] For a general discussion of the theory of EELS in a layered system, see D. L. Mills *et al.,* Phys. Rev. B **50**, 6394 (1994). Note that the experiments analyzed in this paper are at energies well below those of interest in the present context.

[6] N. Nucker *et al.,* Phys. Rev. B **39**, 12 379 (1989); Y. Y. Wang *et al., ibid.* **42**, 420 (1990).

[7] It is straightforward to obtain this formula case by case for $n \leq 6$ and $n = \infty$. I am indebted to Misha Turlakov for a proof of its general validity.

[8] A more careful treatment of the cutoff [2] actually multiplies the right-hand side of Eq. (6) by a factor of approximately 2.

[9] Any failure of assumptions (A4) and/or (A7) is likely if anything to strengthen this conclusion [2].