sponsible for the effect. Excitation energy transfer between the D_1 and D_2 chromophores (leading to exciton annihilation if the two are simultaneously excited) can also be ruled out by observation of the evolution of the transient absorption spectra when using only a single pump. Selective excitation of each donor in D_1-A_1-A_2-D_2 (D_1 at 416 nm, D_2 at 512 nm) leads to the selective appearance of the anion spectrum of the acceptor adjacent to the donor that is excited. The anion absorption spectra of the two acceptors are readily distinguishable due to their sharp and distinctive features. This observation also allows us to rule out the possibility of electron transfer from D_1-A_1-A_2^--D_2^+ to D_1-A_1^--A_2-D_2^+. This process, although energetically favorable, is presumably too slow to compete with recombination of A_2^--D_2^+ to the ground state.

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Interlayer Tunneling Models of Cuprate Superconductivity: Implications of a Recent Experiment

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It is shown that, given a few generic assumptions, any theory of high-temperature superconductivity that attributes a substantial fraction of the condensation energy to the saving of *c*-axis kinetic energy must predict an inequality relating the *c*-axis penetration depth $\lambda_\perp(0)$ to the zero-temperature superconducting-normal energy difference and the fluctuations of the *c*-axis kinetic energy around its mean value. Application of this formula to $Tl_2Ba_2CuO_6$ implies that if $\lambda_\perp(0)$ is greater than 10 micrometers, as suggested by a recent experiment, these fluctuations must have an unusual form.

Of the many and varied approaches to the high-temperature superconductivity (HTS) problem currently available, none is more novel or intriguing than the interlayer tunneling (ILT) model of Anderson and coworkers (*1–4*). This model rests on two major postulates: (i) The normal state of the electrons within a single CuO_2 plane is different in nature from the traditional Landau Fermi liquid, and (ii) as a result, single-particle tunneling between the CuO_2 planes is strongly inhibited in the normal phase; however, in the superconducting phase, tunneling of pairs is possible and results in a strong decrease of the *c*-axis kinetic energy $T_\perp$. Thus, depending on the particular version of the model considered, either all or, at least in the cuprates with higher transition temperatures T_c, a large fraction of the condensation energy E_{cond} (the difference between the superconducting ground-state energy and the "best" normal-state energy) comes from the decrease of $\langle T_\perp \rangle$ (brackets denote expectation value) in the superconducting state. In this report, I shall take this last statement as the definition of the ILT model, irrespective of the way in which point (i) is implemented.

The ILT model has a number of attractive features. It explains in a natural way why all materials known to date with $T_c >$ 35 K have well-separated planes in which the electronic behavior appears to be two-dimensional (2D) in the first approximation, and why despite the apparent similarity of the in-plane behavior, the values of T_c for the various cuprates vary by a factor of more than 20. The model obviates the need for an anomalously strong in-plane attraction mechanism, and it can be argued (*2*) to be consistent with a variety of apparently puzzling experimental properties of the cuprates, in particular (*4*) with the normal- and superconducting-state finite-frequency *c*-axis conductivity. It is therefore important to explore the generic consequences of the ILT model and to examine their compatibility with existing experimental data.

For this purpose, I confine myself to those ("single-plane") cuprates in which all pairs of neighboring CuO_2 planes are equivalent. In this case, various particular implementations of the ILT model [see especially (*3*)] lead to an expression for the $T = 0$ interplane coupling energy E_{int} (which in these versions is the difference between the normal- and superconducting-state values of $\langle T_\perp \rangle$) that is a special case of the more general Lawrence-Doniach (LD) expression (given for a particular pair of planes i, $i + 1$)

$$E_{int}^{LD} = K - J\cos\Delta\phi_i^{(pair)} \tag{1}$$

where $\Delta\phi_i^{(pair)}$ is the difference in the phase of the superconducting order parameter between the two planes in question, and K and J are material-dependent constants. In non-ILT theories, which simply regard the interplane tunneling process as described by the original Josephson model (*5*) for a tunnel oxide barrier and naïvely apply the Ambegaokar-Baratoff expressions (*6*), one finds that $K \approx J$, and hence in the superconducting ground state [$\Delta\phi_i^{(pair)} = 0$], no appreciable energy is saved by tunneling. However, in the current form of the ILT model (*3, 4*), K is postulated to be zero, or at any rate, $K \ll J$. If this is so, one obtains a relation between the saving of *c*-axis kinetic energy in the superconducting ground state, $\Delta T_\perp^{ns}$, and the $T = 0$ *c*-axis superfluid density $\rho_{s\perp}$: for $K = 0$

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

where m is the electron mass and d is the interplanar spacing. In the ILT model, $\Delta T_\perp^{ns}$ supplies a substantial fraction η of the superconducting condensation energy, which, as pointed out by Anderson (*4*), leads to a definite prediction for the $T = 0$ *c*-axis penetration depth $\lambda_\perp(0)$. For our purposes, it is convenient to write this prediction in the form

$$\lambda_\perp = \frac{1}{2}\eta^{-1/2}\lambda_{ILT}, \quad \lambda_{ILT} = \left(\frac{mc^2}{E_{cond}}\frac{a_0 A}{4\pi d}\right)^{1/2} \tag{3}$$

where c is the speed of light, E_{cond} is the superconducting condensation energy per formula unit, a_0 is the Bohr radius, and A is the area per formula unit. (The precise definition of λ_{ILT} is chosen with a view to giving Eq. 7 below a simple form.) A case of particular interest is $Tl_2Ba_2CuO_6$ (Tl-2201), where $\lambda_{ILT} \sim 1.8$ μm. Recently, van der Marel *et al.* (*7*) concluded, on the basis of the absence of a *c*-axis plasmon peak in their optical reflectivity measurements, that $\lambda_\perp(0)$ must be >10 μm. From Eq. 3, this implies that $\eta < 0.01$ in this material; this conclusion holds for any positive value of K in Eq. 1.

A value of $\lambda_\perp(0) > 10$ μm in Tl-2201, if confirmed, would thus seem to refute definitively any version of the ILT model, such

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as that of (*3, 4*), in which $\Delta T_\perp^{ns}$ is given by (the negative of) Eq. 1 with $K \geq 0$. However, it may be argued that there is no particular reason why a generic version of the model should lead to Eq. 1, or even if it does, why K cannot be negative. Thus, it is of interest to inquire whether there are any general constraints that a large value of $\lambda_\perp(0)$ would put on the ILT model.

The purpose of this report is to point out observations. (i) Although in a periodic system there is no generic formula (*8*) for the ratio of the expectation value of $T_\perp$ in the superfluid ground state, $\langle T_\perp \rangle_0$, to the quantity $\rho_{s\perp}$, it is possible to find an upper bound on this ratio in terms of a dimensionless parameter. The parameter is a measure of the probability of anomalously small values of $T_\perp$. (ii) Large values of $\lambda_\perp(0)$ thus imply large values of this parameter.

To be specific, I consider a perfect single-plane cuprate, described by a Hamiltonian $\hat{H}$ and a (superconducting) ground state Ψ_0 such that the following rather weak set of conditions is satisfied.

1) $\hat{H}$ is of the form $\hat{H}_0 + \hat{T}_\perp$, where $\hat{H}_0$ contains in-plane kinetic energies and (intra- and interplane) electron-electron interactions, ion kinetic energies and electron-ion interactions, and is invariant under time reversal, and $\hat{T}_\perp$ is a simple one-parameter tight-binding Hamiltonian for the interplane motion, with a parameter $t_\perp$ that may depend on the ionic coordinates (and possibly also on the coordinates of the electrons other than the hopping one).

2) The ground state Ψ_0 does not spontaneously break the crystalline or time-reversal invariance of $\hat{H}$.

3) The real part of the *c*-axis conductivity $\sigma(\omega, T)$ tends to zero as $T \to 0$ and the frequency $\omega \to 0$.

4) I also impose for the moment a more problematic condition: The quantity $T_i(\xi)$ defined below in Eq. 4 is positive definite except for a set of ξ of measure zero (*9*).

Let the integer $z_k = 1, 2, \ldots, n$ denote the plane (layer) index of the *k*th electron ($k = 1, 2, \ldots, N$), and let ξ stand for all coordinates of the system other than z_1. Recall that Ψ_0 is real because time-reversal invariance is not broken, and define

$$T_i(\xi) \equiv 2t_\perp(\xi)[\Psi_0(z_1 = i, \xi)\,\Psi_0(z_1 = i + 1, \xi)] \qquad (4)$$

The crucial point in the ensuing argument is that, despite condition (2), for given ξ, the quantity $T_i(\xi)$ may depend on i, and therefore, the "best" wave function may for given ξ have a different phase drop across different pairs i, $i + 1$.

Then I assert that, subject only to the above conditions, the following inequality holds

$$-\langle T_\perp \rangle_0 \leq \alpha\left(\frac{\hbar}{md}\right)^2 \rho_{s\perp} \qquad (5)$$

where

$$\alpha \equiv \lim_{n\to\infty} \frac{\langle T_i \rangle}{n\left\langle \left(\sum_{i=1}^{n} T_i^{-1}\right)^{-1} \right\rangle} \qquad (6)$$

Equation 5 is actually a special case of a more general inequality for the superfluid density of a many-body system; here I sketch the bare bones of the derivation. (i) From postulate (*3*), the system can only respond adiabatically to the "phase twist" $\Delta\phi$ in the (single-particle) boundary condition that defines the superfluid density, and from postulates (1) and (2), the deformation in the limit $\Delta\phi \to 0$ must consist of simply multiplication by a phase factor $\exp[i\Phi(z_1, \xi)]$, which must satisfy, among other things, the boundary condition $\Phi(z_1 = n, \xi) - \Phi(z_1 = 1, \xi) = \Delta\phi$ for all ξ. (ii) The increase in $\langle T_\perp \rangle$ is a lower bound (in the limit $\Delta\phi \to 0$) on the increase in $\langle H \rangle$; the former, being given in this limit by $\Sigma_i T_i(\xi)[\Delta\phi_i(\xi)]^2$, where $\Delta\phi_i(\xi) \equiv \Phi(z_1 = i + 1, \xi) - \Phi(z_1 = i, \xi)$, is minimized, subject to the above boundary condition, by the choice $\Delta\phi_i(\xi) \propto [T_i(\xi)]^{-1}$, and the resulting value is simply proportional to $\{\Sigma_i[T_i(\xi)]^{-1}\}^{-1}$. Integration over ξ and use of the standard definition of $\rho_{s\perp}$ then yields the inequality in Eq. 5 (*10*). It should be strongly emphasized that Eq. 5 is an upper bound only and, in particular, it is perfectly possible for α to be infinite without $\rho_{s\perp}$ being zero.

Now it is clear that the value of $\langle T_\perp \rangle$ in the normal ground state cannot be positive (were it so, we could simply perform a gauge transformation to make it negative without affecting $\langle H_0 \rangle$), and thus, the right-hand side of the Eq. 5 also constitutes an upper bound on ΔT_{ns}. Consequently, any theory of the ILT type that attributes a fraction η of the superconducting condensation energy to ΔT_{ns} must predict the inequality

$$\alpha \geq \eta \frac{\lambda_\perp^2(0)}{\lambda_{ILT}^2} \qquad (7)$$

where $\lambda_\perp(0)$ is measured experimentally.

Equation 6 shows that α is a measure of the fluctuations of $T_i(\xi)$ around its mean value, or more precisely, of the likelihood of occurrence of very small values of (special elements of) this quantity. In the present context, the interesting question is how plausible is a large value (say $\gtrsim 30$) of α? Defining t_i as the ratio of T_i (per formula unit) to the value of the hopping matrix element $t_\perp$ calculated by band-theory techniques, we can write $\alpha \leq \bar{t}\cdot\overline{t^{-1}}$, where I use the inequality, valid for any positive definite function χ, $\langle \chi^{-1} \rangle \geq \langle \chi \rangle^{-1}$. It follows that if the distribution $f(t)$ behaves in the limit $t \to 0$ as $t^{-(1+\gamma)}$ (with a lower cutoff if $\gamma > 0$), then a value of α greater than 30 implies that γ must lie either above -1 or very close to it (the precise range being determined by the details of the larger $-t$ behavior). So far, I believe that the results are rigorous.

Physical intuition tells us, however, that Eq. 5, with Eq. 6, is in reality too weak a lower bound on the superfluid density. Consider, for example, the effect of one particular variable entering the set $\{\xi\}$, namely, the in-plane coordinate of the *i*th (tunneling) electron. The reason that α in Eq. 6 can be large, even infinite, is simply that in a strictly 1D system, the probability of finding an arbitrarily weak link tends to unity as $N \to \infty$. In reality, the supercurrent "shunts" any such weak links by flowing sideways in the plane until it finds a stronger one. Thus, to have an "effective" link of strength less than t, it is necessary (although far from sufficient) for this value to apply over an area of the order of L^2, where L is such that $(\hbar^2/2mL^2)(\rho_s^{ab}/\rho) \sim tt_\perp$ and ρ_s^{ab} is the superfluid density in the *ab* plane. Let us define the parameter $\kappa \equiv (\hbar^2/2ml_0^2 t_\perp)(\rho_s^{ab}/\rho)$, where l_0 is the correlation length of t within the plane. Because the correlations are likely to be themselves primarily determined by in-plane rather than interplane effects, it seems likely that κ is large compared to 1; in fact, for most realistic models it should be of order of the anisotropy $t_\perp/t_{ab}$ of the tight-binding matrix elements (probably ~50 for Tl-2201). The number of independent links that must be in parallel is at least of order κt^{-1}, and a fairly straightforward combinatorial argument shows that the "effective" value of α so produced, for negative γ, is such that $\alpha\kappa|\gamma|(\ln \alpha) \lesssim 1$; thus, for $\alpha \gtrsim 30$, the quantity γ, if negative, must be exceedingly small. For positive γ, the constraint is weaker: If the lower cutoff is ε_c, then the relevant inequality is $\kappa\alpha(\varepsilon_c\alpha)^\gamma \lesssim 1$. However, because in the ILT model the quantity $\bar{t}$ is constrained to be at least $E_{cond}/t_\perp$, a ratio that I estimate (*11*) at about 5×10^{-3}, it follows that unless γ has a large value ($\gtrsim 1.7 + 0.5\ln \kappa$), $\bar{t}$ cannot be determined by the asymptotic form of the fluctuations but must have substantial contributions from larger values of t. The most "naïve" model for producing large probabilities of small values of t—albeit one that is arguably not in the spirit of the ILT concept, namely a model in which (i) the relative energy ΔE of neighboring planes fluctuates with a root-mean-square amplitude large compared to $t_\perp$ and (ii) $t \sim t_\perp^2(\Delta E^2 + t_\perp^2)^{-1/2}$ (so that $\gamma = 1$)—does not satisfy the constraints derived here [which are necessary but by no means sufficient for the viability of the ILT model in the face of a large value of $\lambda_\perp(0)$].

With the above analysis in mind, let us consider the consequences of relaxing condition (4). The argument leading to Eq. 5 now fails because it may be possible to put in a much larger than average value of $\Delta\phi_i$ across links for which $T_i(\xi)$ is negative, thereby actually decreasing $\langle T_\perp \rangle$ by itself. However, consider a set of $\mathcal{N}$ links with an average value of $T_\perp(\xi)$ equal to $-\varepsilon\langle T \rangle$; then an (not entirely trivial) argument analogous to that of the proceeding paragraph shows that for such a configuration to give an energy advantage, we must have $\mathcal{N} \gtrsim \kappa/\varepsilon\bar{t}$ (or $\kappa/\varepsilon^2\bar{t}$ for $\varepsilon \lesssim 1$). Bearing in mind that by definition $\varepsilon\bar{t} \lesssim 1$ and that we have estimated $\kappa \sim 50$, we now ask if it is possible to find a distribution $f(t)$ of the $T_i(\xi)$—even a pathological one—such that the probability of such an occurrence is, as a minimum, comparable to the inverse of the number of planes ($\sim 10^{-7}$, say). I have so far failed to find such a distribution and, although I have at present no rigorous proof, strongly suspect that none exists. Thus, although relaxation of condition (4) at first appears to prevent the derivation of a rigorous inequality such as Eq. 5, it should not affect the qualitative conclusions reached above.

The conclusion is that although a value of $\lambda_\perp(0)$ for Tl-2201 greater than 10 μm would not definitively refute a generic form of the ILT model, it would as a minimum either violate one or more of assumptions (1) through (3) or constrain the ground state to have extremely large fluctuations in $T_\perp$ (which should, at least in principle, be detectable in angle-resolved photoelectron spectroscopy measurements on *a*- or *b*-axis–oriented films). It remains to be seen whether a concrete version of the model having this feature can be constructed and shown to be physically reasonable.

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8. The reader might reasonably ask why we should not simply use equation 1 of (*12*) and use the experimental ac conductivity to evaluate the second term. Although this is in principle possible, in practice any analysis in the cuprate superconductors that relies on finite-frequency effects is fraught with notorious difficulties. The advantage of the present analysis is that it relies only on properties of the ground state [note that there is no reason why $\lambda_\perp(0)$ itself should not be measured by a dc technique].
9. The function $T_i(\xi)$ cannot be positive definite for all ξ because of the requirement of Fermi antisymmetry. Assumption (4) is in effect the statement that the antisymmetry is completely satisfied by the in-plane behavior, and it turns out not to be satisfied by some familiar models, for example, a weakly paired 3D Fermi liquid with a tight-binding normal-state spectrum in the *c* direction; note however that it is satisfied by, for example, the standard LD model, provided that the matrix element $t_\perp$ is constant across the *ab* plane.
10. It may be objected that (the analog of) Eq. 5 is spectacularly violated for the standard [Ambegaokar-Baratoff (*6*)] model of a single Josephson junction (which at first sight corresponds to the special case of the above calculation for $n = 2$) because it is a standard result that the Josephson coupling energy (the "superfluid density" up to a factor) is a small fraction of the total tunneling energy, whereas the quantity α is of order unity. However, to analyze this model along the above lines, one must consider the junction itself as being in series with whatever mechanism is responsible for "dephasing" the electrons in the bulk superconductors, as well as the effect of (non)conservation of the transverse momentum. Thus, it is not (or at least not obviously) a counterexample to the theorem (*3*).
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Evidence for Charge-Flux Duality near the Quantum Hall Liquid-to-Insulator Transition

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A remarkable symmetry has been observed between the diagonal, nonlinear, current-voltage (I-V_{xx}) characteristics taken in the fractional quantum Hall effect (FQHE) liquid state of the two-dimensional electron system and those taken in the bordering insulating phase. When properly selected, the I-V_{xx} traces in the FQHE regime are identical, within experimental errors, to V_{xx}-I traces in the insulator, that is, with the roles of the currents and voltages exchanged. These results can be interpreted as evidence for the existence of charge-flux duality symmetry in the system.

The theoretical understanding of the quantum Hall effect (QHE) is believed to involve the physics of the ideal states and that of localization. A precise theoretical account of the QHE phenomena is, not surprisingly, controversial, for it requires solving a problem with interactions, fractional statistics, and disorder—a rather formidable task. Nevertheless, considerable theoretical progress had been made in recent years in understanding the phase diagram of QH states and the transitions between them.

In a recent theoretical paper, Kivelson, Lee, and Zhang (*1*) used a flux attachment (Chern-Simons) transformation (*2*) to map the two-dimensional electron system (2DES) at high magnetic field (B) onto a bosonic system under a different field B_{eff}. The advantage of this mapping is clear if one considers the "magic" Landau-level filling fractions (ν values) where the fractional quantum Hall effect (FQHE) liquid states are observed. At these ν values the Chern-Simons gauge field cancels, on average, the externally applied B, and the composite bosons (CBs) experience a vanishing B_{eff}. The incompressible FQHE states then arise as a result of the formation of a Bose-condensed, superconducting state of the CBs.

At ν values other than the magic ν values, the cancellation of the external B is not exact and, according to the bosonic picture, vortices are created in the CB condensate. For small deviations from magic ν values, the vortex density is small, and the vortices are localized by disorder and do not contribute to the long-wavelength electrical response. When the deviation from the magic ν values becomes sufficiently large, the superconductivity of the CBs is destroyed by the excess magnetic field and the

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