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LETTER TO THE EDITOR

Rosencrantz and Guildenstern may not be dead; on the interlayer Josephson vortices in Tl-2201

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Abstract. Interpreting their experimental data in terms of an approximate solution of the Lawrence–Doniach–Clem (LDC) model in the continuum limit (describing an isolated interlayer Josephson vortex), Moler *et al* (1998 *Science* **279** 1193) have estimated the penetration depth λ_c in the direction normal to the layers of the compound $Tl_2Ba_2CuO_{6+\delta}$ (Tl-2201) to have the value ≈ 22 μm. They thus concluded that only $\approx 0.1\%$ of the superconducting condensation energy in Tl-2201 would be due to the interlayer-tunnelling mechanism. We have studied the LDC model and found that it has *no* physical solution. Therefore λ_c in Tl-2201 is in need of re-examination.

Within the framework of the interlayer tunnelling (ILT) theory [1–3] for superconducting cuprates with high transition temperatures (high-T_c superconductors), the mechanism of Cooper pairing is at work. This pairing is brought about by the gain in the free energy of the system associated with the process of *coherent* interlayer tunnelling of the *paired* electrons, a process which is assumed to be *incoherent* for unpaired electrons. This discriminating effect is ascribed to the non-Fermi liquid nature of the low-energy excitations in the normal states of high-T_c cuprates; explicitly, these excitations are not particle-like, unlike the Landau quasiparticles, but collective. According to the ILT scenario, the superconducting condensation energy is (almost) *entirely* equal to the Josephson-coupling energy. Since the latter energy is inversely proportional to the square of the c-axis penetration depth λ_c, the superconducting condensation energy and consequently the superconducting transition temperature T_c must rapidly decrease for increasing values of λ_c. Therefore for the ILT mechanism to provide a viable explanation for the phenomenon of high-T_c superconductivity in the known cuprates, it is necessary that λ_c in these compounds be on the order of 1 μm (however, see [4]). For the high-T_c compounds $(La, Sr)_2CuO_4$ ('214') and $HgCa_2CuO_4$ (Hg-1201), λ_c amounts to respectively $\approx$3 and $\approx$1 μm [3].

Moler *et al* [5] have recently measured the magnetic flux due to some isolated interlayer Josephson vortices in samples of the single-layer compound $Tl_2Ba_2CuO_{6+\delta}$ (Tl-2201) by means of a small pick-up loop moved at constant heights ($z_0 \approx 3$ μm) from surfaces of these parallel to the ac-plane (the a- and b-axes are normal to the c-axis and thus span a plane parallel to the CuO planes). Through a fitting procedure (for details see further on) involving the measured magnetic fluxes as functions of position of the pick-up loop with respect to locations of some well-isolated interlayer vortices, these authors have determined $\lambda_c \approx 22$ μm for Tl-2201. This large value for λ_c implies that (i) the ILT mechanism is not operative in Tl-2201 and, most importantly, (ii) this mechanism is *not* generic for the high-T_c superconductivity in the cuprates.

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For deducing the value of λ_c, Moler *et al* [5] rely on the assumption that the magnetic flux density $B_{\hat{z}}(x, y, z)$ in the z-direction (i.e. the direction coinciding with that of the b-axis) at $z = 0$, the position of the surface of the sample, is *identical* to $b_{\hat{z}}(x, y)$ pertaining to the infinite, surface-free, sample; whence the z-independence of $b_{\hat{z}}(x, y)$. Within the 'elliptical approximation', developed by Clem [6] and Clem and Coffey [7] and employed by Moler *et al* [5] in their analyses, $b_{\hat{z}}(x, y)$ is replaced by $\overline{b}_{\hat{z}}(x, y)$ which is related to $\tilde{b}_{\hat{z}}(\varrho)$ whose significance we shall elaborate upon below. We have

$$\overline{b}_{\hat{z}}(x, y) := \frac{\Phi_0}{2\pi s \lambda_c} \tilde{b}_{\hat{z}}\big(([x/\lambda_a]^2 + [y/\lambda_c]^2)^{1/2}\big) \tag{1}$$

where

$$\tilde{b}_{\hat{z}}(\varrho) \equiv \frac{2\pi s \lambda_c}{\Phi_0} b_{\hat{z}}(x = 0, \lambda_c \varrho). \tag{2}$$

Here $\Phi_0 := hc/[2e]$ stands for the superconductor magnetic-flux quantum with h the Planck constant, c the speed of light in vacuum and $-e$ (< 0) the electron charge; λ_a denotes the in-plane penetration depth (assumed to be isotropic, i.e. $\lambda_a = \lambda_b$) and s the sum of the superconductor layer thickness d_s and the insulating layer thickness d_i. In this work we shall follow Moler *et al*, [5] and for s and λ_a, pertaining to Tl-2201, adopt $s = 11.6$ Å and $\lambda_a = 0.17$ μm. As is evident from equations (1) and (2), for $x \to 0$, $\overline{b}_{\hat{z}}(x, y) \to b_{\hat{z}}(0, y)$.

The magnetic flux density $b_{\hat{z}}(x, y)$ is determined from a set of two coupled non-linear partial differential equations (DEs) which involve the gauge-invariant Josephson phase-difference $\Delta\gamma_n(x, y)$ corresponding to the nth and the $(n + 1)$th superconducting layers. Specializing to the *central* layer, corresponding to $n = 0$ and $x = 0$ (the point $x = y = 0$ coincides with the core centre of the isolated Josephson vortex), Clem and Coffey [7] have obtained a coupled set of non-linear *ordinary* DEs for $b_{\hat{z}}(0, y)$, which, in consequence of equation (2), give rise to the following equations [8] for $\tilde{b}_{\hat{z}}(\varrho)$ and $\phi(\varrho) := \Delta\gamma_0(x = 0, \lambda_c \varrho)$:

$$\tilde{b}_{\hat{z}}(\varrho) = -\frac{\partial \phi(\varrho)}{\partial \varrho} - \frac{\sin \phi(\varrho)}{\varrho} \tag{3}$$

$$\frac{\partial^2 \phi(\varrho)}{\partial \varrho^2} + \frac{1}{\varrho} \frac{\partial}{\partial \varrho} \sin \phi(\varrho) - \left(1 + \frac{1}{\varrho^2}\right) \sin \phi(\varrho) = 0. \tag{4}$$

A third equation, associated with the Josephson relation, from which, in combination with equation (3), equation (4) has been obtained, reads

$$\frac{\partial \tilde{b}_{\hat{z}}(\varrho)}{\partial \varrho} = -\sin \phi(\varrho). \tag{5}$$

The physical solution $\phi(\varrho)$ of equation (4) has to satisfy the boundary conditions (BCs) $\phi(0) = \pi$ and $\phi(\varrho \to \infty) \to 0$ [7]. In addition to these, the fluxoid-quantization condition (FQC)

$$\int_0^\infty \mathrm{d}\varrho \varrho^2 \sin \phi(\varrho) = \frac{2s}{\lambda_a} \tag{6}$$

has to be fulfilled [7]. This latter condition follows from $\int_0^\infty \mathrm{d}\varrho \varrho\, \tilde{b}_{\hat{z}}(\varrho) = s/\lambda_a$, which in turn is a consequence of $\int \mathrm{d}x \mathrm{d}y b_{\hat{z}}(x, y) = n\Phi_0$ corresponding to $n = 1$ (the case of the singly-quantized fluxoid) and equations (1) and (2). The condition $\phi(0) = \pi$ is due to the reflection symmetry of the model with respect to the zx-plane which, because of phase symmetry [9], implies $\Delta\gamma_0(0, y) = 2\pi - \Delta\gamma_0(0, -y)$, whereas $\phi(\varrho \to \infty) \to 0$ is required for the total magnetic flux due to a vortex to be finite; as is evident from equation (6), for this $\phi(\varrho)$ has to approach zero not slower than $1/\varrho^p$ with $p > 3$. The same requirement implies that for $\varrho \to \infty$, $\tilde{b}_{\hat{z}}(\varrho)$ has to vanish not slower than $1/\varrho^p$ with $p > 2$. Similarly, for $\varrho \downarrow 0$, $\tilde{b}_{\hat{z}}(\varrho)$ must not diverge faster than $1/\varrho^p$ with $p \geqslant 2$. An asymptotic analysis of equation (4) yields

$$\phi(\varrho) \sim \pi - \Big(C_0'\varrho\, J_0(\varrho) + C_0''\varrho\, Y_0(\varrho)\Big) \sim \pi - \Big(C_0'\varrho + \frac{2}{\pi}C_0''\varrho\,\ln(\varrho)\Big) \qquad \varrho \downarrow 0 \tag{7}$$

$$\phi(\varrho) \sim C_\infty K_1(\varrho) \sim C_\infty \left(\frac{\pi}{2\varrho}\right)^{1/2} \exp(-\varrho) \qquad \varrho \to \infty. \tag{8}$$

Here J_ν, Y_ν and K_ν stand, respectively, for the order-ν Bessel functions of the first kind, the second kind and the modified third kind [10]; C_0', C_0'' and C_∞ are constants whose method of determination we shall describe further on in this work. Equation (7) implies that $\partial\phi(\varrho)/\partial\varrho$ is logarithmically divergent at $\varrho = 0$. From equations (3) and (7) it readily follows that for $\varrho \downarrow 0$, $\tilde{b}_{\hat{z}}(\varrho) \sim 2C_0''/\pi$; for $\varrho \to \infty$, equations (3) and (8) yield $\tilde{b}_{\hat{z}}(\varrho) \sim C_\infty(\pi/[2\varrho])^{1/2}\exp(-\varrho)$. Both of these results are consistent with the above-indicated requirements to be met by $\tilde{b}_{\hat{z}}(\varrho)$ for the flux due to a Josephson vortex to be finite.

It is interesting to note that the physical solution $\phi(\varrho)$ of equation (4) is *independent* of λ_c: all functions in equation (4) are universal, as are the boundary values $\pi = \phi(0)$ and $0 = \phi(\infty)$. Further, $\phi(\varrho)$ merely parametrically depends on the physical quantities s and λ_a, via the FQC in equation (6), and in the particularly limited way that only the *ratio* s/λ_a is of influence. The sole way in which λ_c enters on the scene is through setting the unit of length according to which y is measured in terms of ϱ. These observations clearly underline the *a priori* limitations of the 'elliptical approximation' (see also text following equation (10) below). In fact these observations gave us the initial impetus to investigate the problem at hand in some detail.

Below we shall argue that the DE in equation (4) together with the mentioned BCs and the FQC amount to a *physically* over-specified problem. It turns out that as a consequence of this, the problem at hand has *no* physical solution. This over-specification is partly due to the above-indicated approximation concerning $b_{\hat{z}}(x, y)$ in the central layer, restricting $b_{\hat{z}}(x, y)$, a function of two variables, to $\tilde{b}_{\hat{z}}(\varrho)$, one of a single variable, and partly due to ϱ being limited to the form $\varrho \equiv \varrho(x, y) = ([x/\lambda_a]^2 + [y/\lambda_c]^2)^{1/2}$, confining the contours on the xy-plane corresponding to constant values of $\tilde{b}_{\hat{z}}(\varrho)$ to be ellipses.

The approximate 'solution' put forward by Clem and Coffey [6, 7] for $\phi(\varrho)$—which we shall denote by $\phi_C(\varrho)$—and employed in the analyses by Moler *et al* [5], has *functionally* the same form as the asymptotic expression in the second term on the right-hand side of equation (8), however, it involves a modified argument which approaches ϱ for large values of ϱ. We have

$$\phi_C(\varrho) := \frac{s}{\lambda_a} K_1\big((\eta^2 + \varrho^2)^{1/2}\big) \tag{9}$$

where $\eta \equiv s/[2\lambda_a]$. A non-vanishing η, irrespective of its precise value, serves to prevent the corresponding magnetic flux density $\tilde{b}_{\hat{z}}(\varrho)$, i.e. $\tilde{b}_{\hat{z};C}(\varrho) \equiv (s/\lambda_a)K_0\big((\eta^2 + \varrho^2)^{1/2}\big)$, from being unbounded at $\varrho = 0$. We point out that $\phi_C(0) \sim 2$, for $(s/\lambda_a) \downarrow 0$, in violation of the BC, namely $\phi(0) = \pi$. This demonstrates that $\phi_C(\varrho)$, in contrast to the prevailing view [6, 7], *cannot* be a variational approximation to $\phi(\varrho)$. Aside from this, the logarithmic divergence of $\partial\phi(\varrho)/\partial\varrho$ $(\sim -(2/\pi)C_0''\ln(\varrho) - (C_0' + 2C_0''/\pi))$ for $\varrho = 0$ signifies that violation of $\phi(0) = \pi$ by $\phi_C(\varrho)$ leads to a substantial deviation of $\phi_C(\varrho)$ from its 'exact' counterpart (assuming an exact counterpart existed) in the close vicinity of $\varrho = 0$; from equation (3) this deviation is seen to lead to a considerable deviation of $\tilde{b}_{\hat{z};C}(\varrho)$ from its 'exact' form (again, assuming that an exact form existed); see [11].

The value $\eta = s/[2\lambda_a]$ has been derived by Clem and Coffey [7] through requiring a matching (applicable in the region $\varrho \gg \eta$) between $(s/\lambda_a)K_0(\varrho)$ for small values of ϱ, so that $K_0(\varrho) \sim -\ln(\varrho)$, with a $\tilde{b}_{\hat{z}}(\varrho)$ as determined from an explicit asymptotic solution of equations (3) and (5) for $\varrho \downarrow 0$ (see [11]). Since for a finite η, $K_1\big((\eta^2+\varrho^2)^{1/2}\big)$ is regular for all ϱ, owing to the ϱ^2 in the integrand of equation (6) combined with the exponential decay of $K_1(\varrho)$ for large ϱ, to a good approximation $\sin\big([\eta/2]\phi_C(\varrho)\big)$ can, *under the integral sign*, be replaced by $[\eta/2]\phi_C(\varrho)$, which, following the exact result $\int_0^\infty \mathrm{d}\varrho\,\varrho^2 K_1(\varrho) = 2$, leads to $\phi_C(\varrho)$ to an extremely good approximation satisfying the FQC in equation (6); for $s/\lambda_a \lesssim 10^{-2}$, $\tilde{b}_{\hat{z};C}(\varrho)$ violates the FQC by not more than five parts in 10^3 (see $I_\beta(0)$ below; note that $\eta_{\mathrm{Tl-2201}} \approx 3.4 \times 10^{-3}$).

The above details serve to specify the degree to which $\phi_C(\varrho)$ can be considered to 'satisfy' equation (4), the BCs and the FQC.

We have attempted to solve the non-linear DE in equation (4) numerically, taking into account the appropriate BCs and the FQC. The BCs involve three undetermined coefficients, C_0', C_0'' and C_∞, which were to be obtained within the following framework: by selecting a (large) value ϱ_0, we have first chosen a dense set of points $\{\rho_i | i = 1, 2, \ldots, M; \rho_i < \rho_{i+1}\}$, ($M \sim 10^3$) over the interval $[0, \varrho_0]$ (in the course of integrating the DE, M could automatically be increased to achieve a specified accuracy in the calculated $\phi(\varrho)$, thus in some cases we have had $M \sim 10^4$ for $\varrho_0 \approx 3$); we have chosen $\{\rho_i\}$ to be the set of shifted and rescaled zeros of the Legendre polynomial [10] of order M, thus ensuring a dense distribution of the ρ_i points close to $\varrho = 0$ and ϱ_0; to exclude $\varrho = 0$ from $\{\rho_i\}$ (as our formulae involve, e.g., $\ln(\varrho)$), we have restricted M to odd integer values. We have subsequently required that (A) $\phi(\varrho)$ as extrapolated to $\varrho = 0$ be equal to π; (B) $\partial\phi(\varrho)/\partial\varrho$ coincide with $C_\infty \partial K_1(\varrho)/\partial\varrho$ at $\varrho = \rho_M$, and (C) the FQC in equation (6) be satisfied (as we shall point out below, these three requirements are not unique). We have employed the following formulation for the FQC: $\int_0^{\varrho_0} \mathrm{d}\varrho\,\varrho^2 \sin\phi(\varrho) = (2s/\lambda_a)\{1 - (\lambda_a/[2s]) \int_{\varrho_0}^\infty \mathrm{d}\varrho\,\varrho^2 \sin\big(C_\infty K_1(\varrho)\big)\}$. This choice, though seemingly trivial, is of fundamental importance owing to the practical limitation as to the maximum interval $[0, \varrho_0]$ over which the DE can be numerically integrated (see further on). Assuming $C_\infty \approx s/\lambda_a \ll 1$ (compare with equations (8) and (9)), the following collection of results for $I_\beta(\varrho_0) := \beta^{-1} \int_{\varrho_0}^\infty \mathrm{d}\varrho\,\varrho^2 \sin\big(\beta K_1(\varrho)\big)$, with $\beta \to 0$ (compare with $2s/\lambda_a \ll 1$), clarify our point of view: $I_\beta(0) \approx 1.996$, $I_\beta(1) \approx 1.623$, $I_\beta(2) \approx 1.014$, $I_\beta(3) \approx 0.553$, $I_\beta(3.5) \approx 0.396$, $I_\beta(4) \approx 0.278$. These show that despite the rapid decrease of $K_1(\varrho)$ for increasing ϱ, a substantial contribution to the right-hand side of the FQC in equation (6) could be due to the asymptotic tail of $\phi(\varrho)$. We mention that for the purpose of evaluating the integral involved in the FQC, we have first constructed a monotonicity-preserving cubic Hermite interpolant from $\{\phi(\rho_i')\}$ on the set $\{\rho_i'\} \supseteq \{\rho_i\}$, i.e. $\{\rho_i\}$ as extended in the course of integrating the DE (see above), and subsequently exactly integrated this interpolant [12].

For a given set of coefficients $\{C_0', C_0'', C_\infty\}$, we have numerically solved the DE in equation (4) by means of the so-called deferred-correction technique combined with a Newton iteration procedure [12]; we have also used a Runge–Kutta–Merson algorithm combined with a Newton iteration within a shooting-and-matching framework [12], obtaining, to the accuracy of our numerical calculations, identical results. With both of these methods one has the possibility of specifying *four* BCs of which *two* must be exact and *two* possibly approximate, subject to the restriction that the exact conditions must not all correspond to the same boundary. In applying these methods, we have first transformed the second-order DE for $\phi(\varrho)$ into a set of two coupled first-order equations by means of introducing the auxiliary function $\varphi(\varrho) := \partial\phi(\varrho)/\partial\varrho$. Of the four conditions at our disposal, concerning $\phi(\varrho)$ and $\varphi(\varrho)$ at $\varrho = \rho_1$ and $\varrho = \rho_M$, we have chosen $\varphi(\rho_1)$ and $\phi(\rho_M)$ as being exactly described by the expressions in equations (7) and (8); in evaluating these expressions we have employed J_0, Y_0 and K_1 rather than their corresponding leading-order asymptotic expressions given on the right-most parts of equations (7) and (8). This choice for the two *exact* BCs clarifies our above requirements (A) and (B). If, for instance, one of the *exact* BCs were $\phi(0) = \pi$, then the above requirement (A) would have to be different. We have numerically examined various alternatives, and the conditions described above have proved to be numerically most robust. Concerning C_0', C_0'' and C_∞, we have attempted to determine these by employing a modification of the Powell hybrid method [12].

Now we present the main observations of our extensive numerical calculations. Before doing so, we should like to emphasize that unless we explicitly indicate otherwise, these observations do *not* constitute mathematical facts, rather they are empirical in that they are outcomes of our extensive and systematic numerical experiments. The utmost care that we have spent in conducting these, however, give us reasoned confidence that our findings are free from numerical artifacts. For completeness, our numerical computations were performed using double-precision arithmetic.

(i) First, there exists *no* $\phi(\varrho)$ that in addition to satisfying the DE in equation (4), conforms with the BCs and the FQC. In other words, there exists no $\{C_0', C_0'', C_\infty\}$ for which the above conditions (A), (B) and (C) can be satisfied. In this connection it is important to mention that by relaxing the condition (C) we obtain a $\phi(\varrho)$, supporting our statement with regard to the over-determinedness of the problem at hand.

(ii) Second, it is possible to construct solutions over $[0, \varrho_0]$ (through numerically integrating the DE, in conformity with the BC at $\varrho = 0$) that at $\varrho_0 \lesssim 3.5$ match the asymptotic tail $C_\infty K_1(\varrho)$ for $\phi(\varrho)$; the union of the two $\phi(\varrho)$s further satisfies the FQC in equation (6). However, in *all* cases investigated by us, these functions have turned out to have unequal slopes at the matching point $\varrho = \varrho_0$.

A discontinuity in the slope of $\phi(\varrho)$ has some far-reaching consequences. For instance, a discontinuity of this type at $\varrho = \varrho_0$ implies $\partial^2\phi(\varrho)/\partial\varrho^2 \propto \delta(\varrho - \varrho_0)$ for $\varrho = \varrho_0$, which can never correspond to a solution of equation (4), as no term in this equation can compensate $\delta(\varrho - \varrho_0)$ to render the left-hand side vanishing [13]. Further, according to equation (3) a discontinuity in the slope of $\phi(\varrho)$ gives rise to a constant contribution to $\tilde{b}_{\hat{z}}(\varrho)$. Since for large values of ϱ, $\phi(\varrho)$ is monotonically decreasing (see equation (8)), such a constant contribution to $\tilde{b}_{\hat{z}}(\varrho)$ can never be compensated by one or a number of possible subsequent discontinuities in the slope of $\phi(\varrho)$. Thus a slope discontinuity in $\phi(\varrho)$, at say $\varrho = \varrho_0 \approx 3.5$, gives rise to $\lim_{\varrho\to\infty} \tilde{b}_{\hat{z}}(\varrho) \neq 0$. This implies violation of the FQC, as presented and discussed in the text following equation (6) above (for $\lim_{\varrho\to\infty} \tilde{b}_{\hat{z}}(\varrho) \neq 0$, equation (6), which has been obtained from the actual FQC through integration by parts, is incomplete and therefore does not show up the problem). To appreciate the drawback of this violation in practical applications, consider the following. For the magnetic flux density at the surface $\mathcal{S}(x, y)$ circumscribed by the pick-up loop, with x and y the Cartesian coordinates of some point of the loop (for the square-shaped loop employed in the experiments, we take this point to coincide with its centre point), Moler *et al* [5] have made use of the electrodynamics of the free space [14], thus interrelating the measured magnetic flux $\Phi_s(x, y, z_0)$ through the pick-up loop to $b_{\hat{z}}(x', y')$ by means of the expression $\Phi_s(x, y, z_0) = \int \mathrm{d}x'\mathrm{d}y' b_{\hat{z}}(x', y') f(x', y'; x, y, z_0)$ where

$$f(x', y'; x, y, z_0) := \frac{z_0}{2\pi} \int_{\mathcal{S}(x,y)} \mathrm{d}x''\mathrm{d}y'' \big((x' - x'')^2 + (y' - y'')^2 + z_0^2\big)^{-3/2}.$$

The former integral is over the entire xy-plane. Within the 'elliptical approximation', employed by Moler *et al* [5] the above expression for Φ_s reduces to

$$\frac{\Phi_s(x, y, z_0)}{\Phi_0} = \frac{\lambda_a}{s} \int_0^\infty \mathrm{d}\varrho\varrho\, \tilde{b}_{\hat{z}}(\varrho)\, g_{\lambda_a,\lambda_c}(\varrho; x, y, z_0) \tag{10}$$

where $g_{\lambda_a,\lambda_c}(\varrho; x, y, z_0) := (2\pi)^{-1} \int_0^{2\pi} \mathrm{d}\varphi f(\lambda_a \varrho \cos(\varphi), \lambda_c \varrho \sin(\varphi); x, y, z_0)$. It is important to notice that the *only* way in which λ_c, within the 'elliptical approximation', exerts influence on the behaviour of Φ_s/Φ_0 is via a linear scaling of one argument of the f-function which is solely characterized by the measuring apparatus (i.e. by $\mathcal{S}$) and the relative position, z_0, of this with respect to some surface; as far as f is concerned, it is immaterial whether the space below this surface is occupied by vacuum or by a superconductor (see our remarks in the paragraph following equation (8) above).

A simple asymptotic calculation reveals that for $\varrho \to \infty$, to the leading order in the inverse of ϱ, the following asymptotic relation holds: $g_{\lambda_a,\lambda_c}(\varrho; x, y, z_0) \sim z_0 \mathcal{A}_{\mathcal{S}} \boldsymbol{E}(1 - \lambda_a^2/\lambda_c^2)/[\pi^2 \lambda_a^2 \lambda_c \varrho^3]$, where $\mathcal{A}_{\mathcal{S}}$ stands for the area of $\mathcal{S}$ and $\boldsymbol{E}(k)$ denotes the complete elliptic integral of the second type [10]; $\boldsymbol{E}(k)$ decreases from $1.57\ldots$ at $k = 0$ to 1 at $k = 1$ so that in view of the smallness of λ_a^2/λ_c^2 in the problem at hand (even for $\lambda_c \approx 1\ \mu$m), to a very good approximation $\boldsymbol{E}(1 - \lambda_a^2/\lambda_c^2)$ in this asymptotic expression can be replaced by unity. Now in view of the ϱ in the integrand in equation (10), a non-vanishing constant for $\tilde{b}_{\hat{z}}(\varrho)$ as $\varrho \to \infty$ leads to a critical dependence of Φ_s/Φ_0 on the value of the upper bound of the ϱ-integration. The value of the coefficient of the $1/\varrho^3$ on the right-hand side of the above asymptotic expression for $\mathcal{A}_{\mathcal{S}} = 8.2 \times 8.2\ \mu\mathrm{m}^2$ (equal to that in the experiments by Moler *et al* [5]), $z_0 = 3\ \mu$m and $\lambda_c = 22\ \mu$m amounts to ≈ 30; for $\lambda_c = 1\ \mu$m this value would be $\approx$700. Suppose now that the ϱ-integration on the right-hand side of equation (10) were carried out over $[0, \varrho_1]$ with $\varrho_1 \geqslant \varrho_0$. From our numerical calculations we have obtained that $(\lambda_a/s)\tilde{b}_{\hat{z}}(\varrho_0) \approx 5.9 \times 10^{-2}$ for $\varrho_0 = 3.5$ (note that $\lambda_a/s \approx 1.5 \times 10^2$) and that $\tilde{b}_{\hat{z}}(\varrho) \approx \tilde{b}_{\hat{z}}(\varrho_0)$ for $\varrho \geqslant \varrho_0$. It trivially follows that by changing ϱ_1 from $\varrho_0 = 3.5$ to ∞, the value of Φ_s/Φ_0 would change with an additive constant, behaving like $1.9 \times (1/\varrho_0 - 1/\varrho_1)$, between 0 and $\approx$0.5. The magnitude of this change in Φ_s/Φ_0 is substantial in view of the fact that the amplitudes of the curves corresponding to Tl-2201 for Φ_s/Φ_0 as measured by Moler *et al* [5] amount to ≈ 0.1. We remark that since for $\varrho \downarrow 0$, $g_{\lambda_a,\lambda_c}(\varrho; x, y, z_0)$ is independent of λ_c (and λ_a), the above asymptotic expression concerning $g_{\lambda_a,\lambda_c}(\varrho; x, y, z_0)$, for large ϱ, makes explicit the crucial role that the tail of $\tilde{b}_{\hat{z}}(\varrho)$ plays in determining Φ_s/Φ_0 and the way in which λ_c affects the latter quantity.

(iii) Last, numerical integration of the DE over $[0, \varrho_0]$ turns out to be extremely difficult to carry out when $\varrho_0 \gtrsim 3.5$; we have not had any success in this for $\varrho_0 > 3.6$. The origin of this difficulty is readily understood by realizing that the contribution of $\phi(\varrho)$ in the tail region $[\varrho_0, \infty)$ to the FQC decreases in significance the larger ϱ_0 becomes (our explicit calculation shows that for $\varrho = 3.5$, this contribution amounts to $\approx 2.3\%$ of $2s/\lambda_a$) so that through increasing ϱ_0, condition (C) turns into the least significant of the three conditions (A), (B) and (C). Consequently, for increasing ϱ_0, the above-discussed slope-discontinuity problem, which we have ascribed to the over-specification of the problem at hand, manifests itself in the 'shrinkage' of the space of functions $\phi(\varrho)$ that, for a *given* $\{C_0', C_0'', C_\infty\}$, satisfy both the DE and the asymptotic boundary conditions in equations (7) and (8). Since the exact $\{C_0', C_0'', C_\infty\}$ has to be obtained iteratively (owing to the non-linearity of the conditions (A), (B) and (C)), for growing ϱ_0 it becomes increasingly more likely that an initial (or trial) choice for $\{C_0', C_0'', C_\infty\}$ renders the conditions in equations (7) and (8) incompatible with the DE: integrating the DE from $\varrho = 0$ upwards and from $\varrho = \varrho_0$ downwards, taking, respectively, the left and right BCs into account, the solutions cannot be matched with a continuous slope. If it turns out that for $\varrho_0 = 3.6$ *all* $\{C_0', C_0'', C_\infty\}$ lead to the above-mentioned incompatibility problem, then it has been rigorously established that 3.6 is very special; with our *finite* number of failed attempts, we are not in a position to assign 3.6 this status, however.

In view of the above considerations, the apparent unavoidability of slope discontinuity in $\phi(\varrho)$ is the clearest indication that the problem at hand is physically ill-imposed. Any approximation, such as $\phi_C(\varrho)$, to the *non-existent* solution of this problem must therefore imply spurious values for some, if not all, of the relevant length scales in the problem at hand. In consequence, the value deduced by Moler *et al* [5] for λ_c concerning Tl-2201 is in serious need of reconsideration.

We are presently attempting to solve the Lawrence–Doniach–Clem model (described by a set of coupled, constrained, non-linear, *partial* DEs) in its general form, bypassing the 'elliptical approximation'. Comparison of the outcomes of these calculations with the experimental results by Moler *et al* [5] will shed light on the value of λ_c as well as other parameters (in particular) in Tl-2201. The non-triviality of this problem is largely due to the fact that these coupled non-linear DEs belong to the class of so-called *stiff* [15] boundary-value problems. The non-separability of the model further enhances the difficulties.

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Note added in proof. The following observations are worth indicating. We have extracted the experimental data for Φ_s/Φ_0 from figure 2(I) in the work by Moler *et al* [5] (120 data points) and attempted to deduce from these various parameters of the system within the framework of the Clem–Coffey [7] 'elliptical approximation'. For an $8.2 \times 8.2\ \mu\text{m}^2$ square-shaped pick-up loop and $\lambda_a = 0.17\ \mu$m (these values are those employed by Moler *et al* [5]), we have obtained $z_0 = 2.97\ \mu$m and $\lambda_c = 22.43\ \mu$m (the standard deviation of our fitted curve from the experimental data for Φ_s/Φ_0 amounts to $\sigma = 4.3\times10^{-3}$), compared with $z_0 = 3.0\pm0.6\ \mu$m and $\lambda_c = 22^{+6}_{-4}\ \mu$m by Moler *et al* [5]. The agreement between the two sets of fitting results give us confidence that our computations have been performed in conformity with those by Moler *et al* [5]. In a series of further computations (based on the same experimental data as above) we have obtained the following results (below by $(a, b, c, \dots; \sigma) = (a', b', c', \dots; \sigma')$ we indicate that a, b, c, etc, have been variables in the fitting process and a', b', c', etc, are the corresponding best values, with $\sigma = \sigma'$ the standard deviation as introduced above; unless indicated otherwise, all lengths are in μm): $(z_0, \lambda_c, \lambda_a; \sigma) = (1.33, 16.79, 6.76 \times 10^{-4}; 3.2 \times 10^{-3})$; $(z_0, \lambda_c, \lambda_a, s; \sigma) = (2.72, 20.62, 3.34 \times 10^{-2}, 170.65$ Å; $3.7 \times 10^{-3})$; $(z_0, \lambda_c, \lambda_a, s, \theta; \sigma) = (2.70, 17.74, 1.62 \times 10^{-2}, 214.87$ Å , $19.89^\circ; 3.2 \times 10^{-3})$; here θ stands for the change in the alignment of the diagonal of the pick-up loop with respect to the y-axis (in other cases where θ is not given, $\theta = 0$). A most striking feature of these results is the substantial underestimation of λ_a with respect to the experimental value, taken to be $0.17\ \mu$m (another striking feature is the overestimation of s by a factor between 15 to 20 as compared with the experimental value of 11.6 Å). As a consequence of this, in all cases we have $s/\lambda_a \sim 1$ which implies violation of the condition for applicability of the Clem–Coffey 'approximation', that is $\eta := s/[2\lambda_a] \ll 1$ (see paragraph containing equation (9); in particular note that for $\eta \not\ll 1$, $\phi_C(\varrho)$ does *not* satisfy the FQC).

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[8] Aside from the 'elliptical approximation' described in the main text, there are two additional approximations involved in the determination of $b_{\hat{z}}(x, y)$ in the bulk of layered superconductors. The first concerns the 'continuum-limit approximation' whereby the differences in the order parameters corresponding to the adjacent superconducting layers in the original Ginsburg–Landau equation, or the Lawrence-Doniach equation (see Tinkham M 1996 *Introduction to Superconductivity* (New York: McGraw-Hill)), are replaced by differentials. Second, for the maximum of the Josephson current $\mathbf{J}_0$ and the penetration depths holds [7]: $\lambda_c^2 = c\Phi_0/[8\pi^2 s\mathbf{J}_0]+(d_s/s)^2\lambda_a^2$ (see the main text). Since $s = d_i + d_s$, we have $d_s/s = 1/[1+d_i/d_s] < 1$, so that in view of $\lambda_c^2/\lambda_a^2 \gg 1$, to a good approximation, $\lambda_c^2 = c\Phi_0/[8\pi^2 s\mathbf{J}_0]$ has been assumed.
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[11] Clem and Coffey [7] define $\alpha(\varrho){:=}\pi - \phi(\varrho)$. Thus from equation (7), $\alpha(\varrho) \sim C_0'\varrho J_0(\varrho) + C_0''\varrho Y_0(\varrho)$, for $\varrho \downarrow 0$. Clem and Coffey obtain however: $\alpha_C(\varrho) \sim 2\arctan(\varrho/\widetilde{\varrho}_0)$, where $\widetilde{\varrho}_0$ is an integration constant, to be compared with our C_0' or C_0''; for $\widetilde{\varrho}_0$, these authors obtain $s/[2\lambda_a]$. This result is *incorrect*, for the reason that Clem and Coffey, in determining the latter asymptotic expression, unjustifiably neglect $\tilde{b}_{\hat{z}}(\varrho)$ in their equation (4.3a) (see the last paragraph in section IV of [7]). This statement is made explicit as follows: for $\varrho \leqslant \widetilde{\varrho}_0$ we have $\alpha_C(\varrho) \sim 2\{\varrho/\widetilde{\varrho}_0 - (\varrho/\widetilde{\varrho}_0)^3/3 + \ldots\}$, whereas $\alpha(\varrho)$ as presented above, owing to the logarithmic singularity of $Y_0(\varrho)$ at $\varrho = 0$, cannot be presented as a Taylor series (asymptotically we have however: $\alpha(\varrho) \sim C_0'\varrho + (2/\pi)C_0''\varrho\ln(\varrho)$). We point out that $\alpha_C(\varrho)$ yields the correct value of 0 for $\varrho = 0$. It is from the above (incorrect) $\alpha_C(\varrho)$ that Clem and Coffey obtain $\tilde{b}_{\hat{z}}(\varrho) \sim -\widetilde{\varrho}_0 \ln\left(\widetilde{\varrho}_0^2 + \varrho^2\right)$, upon Taylor expansion of which for $\varrho/\widetilde{\varrho}_0 \ll 1$ they arrive at $\eta \equiv \widetilde{\varrho}_0 = s/[2\lambda_a]$, whence $\tilde{b}_{\hat{z};C}(0) = (s/\lambda_a)\ln(2\lambda_a/s)$, to be compared with our result $\tilde{b}_{\hat{z}}(0) = 2C_0''/\pi$.
[12] For constructing monotonicity-preserving cubic Hermite interpolants, we have employed the routine E01BEF (this as well as the following are from the *NAG Fortran Library*); for interpolation, based on such an interpolant, we have used the routine E01BFF, and for the exact integration of functions from their cubic-Hermite interpolants, we have relied upon the routine E01BHF. The deferred-correction technique combined with the Newton iteration is performed by the routine D02GAF; the Runge–Kutta–Merson method combined with the Newton iteration technique is performed by the routine D02HAF. For solving the zero-vector $\{C_0', C_0'', C_\infty\}$ we have employed C05NBF and C05NCF, both of which are based on the Powell hybrid method.
[13] Since $\sin(\zeta)$ is an entire function of ζ and both $1/\varrho$ and $1/\varrho^2$ are everywhere, with the exception of $\varrho = 0$, regular, *all solutions of equation (4), irrespective of the BCs or the FQC, are infinitely many times differentiable over* $(0, \infty)$. This follows from the fact that any function satisfying equation (4) must possess a continuous first derivative with respect to ϱ (see text). Differentiating both sides of equation (4) (which is an allowed operation in view of $\phi(\varrho)$ being defined on an *open* interval), one observes that also the second derivative with respect to ϱ of $\phi(\varrho)$ has to be continuous. Proceeding along this line for an arbitrary number of times, our statement is proved. An important corollary to the above statement is that *no non-trivial solution of equation (4) can have a finite support*.
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