# Critical temperature and the Ginzburg–Landau theory of layered high-temperature superconductors

by T. Schneider

**Using the mean-field approximation, we study a model for quasi-two-dimensional superconductors. The interlayer coupling, assumed to be mediated by a small electron-hopping term, is found to leave $T_c$ practically unaffected. Thus, a three-dimensional pairing mechanism is required to explain the observed rise in $T_c$ with decreasing average layer spacing in the Bi and Tl compounds. Taking the inhomogeneities of intrinsic or extrinsic nature into account, we find, in the dirty limit, corrections to the conventional anisotropic Ginzburg–Landau behavior—an upward curvature of the upper critical fields which appears to be a universal feature of layered superconductors.**



## 1. Introduction

Since the discovery of high-temperature superconductivity in the La–Ba–Cu–O system [1], a rapidly growing number of superconducting oxides have been synthesized, with critical temperatures $T_c$ ranging from 5 to 125 K. Particular attention has been attracted by the layered compounds with chemical formula $X_m Ca_{n-1} Ba_2 Cu_n O_{2(n=1)+m}$, where $X$ = Bi or Tl, $m$ = 1 or 2, and $n$ = 1, 2, 3, 4, or 5 [2–10]. These are the striking facts: $T_c$ increases markedly with the number $n$ of CuO sheets per formula unit; properties parallel and perpendicular to the layers are highly anisotropic; layers within a unit are not necessarily equivalent; the transition to the superconducting state is not very sharp, pointing to the importance of inhomogeneities. These properties clearly reveal the layered nature of these compounds. Viewing them as anisotropic superconductors is not sufficient. Such a treatment is valid only when the order parameter varies slowly on the scale of the layer separation, which effectively limits the applicability to an ever-decreasing range of temperatures near $T_c$ as the interlayer coupling is decreased.

We recently presented a microscopic model treating these materials as layered superconductors, allowing inequivalent layers and couplings within a unit cell [11]. Using the mean-

**Table 1** Critical temperature $T_c$ and mean spacing between layers $s = c/q$. $X_m Ca_{n-1} Ba_2 Cu_n O_{2(n+1)+m}$. $q$ denotes the number of layers per unit cell.

| $X$ | $m$ | $n$ | $s$ (Å) | $T_c$ (K) | *Reference* |
|---|---|---|---|---|---|
| Bi | 2 | 1 | 12.3 | 22 | 2 |
| Bi | 2 | 2 | 7.8 | 85 | 6 |
| Bi | 2 | 3 | 6.2 | 110 | 7 |
| Tl | 2 | 1 | 11.6 | 20 | 9 |
| Tl | 2 | 2 | 7.4 | 108 | 9 |
| Tl | 2 | 3 | 6.05 | 125 | 9 |
| Tl | 1 | 1 | 9.7 | 20 | 9, 10 |
| Tl | 1 | 2 | 6.4 | 85 | 9, 10 |
| Tl | 1 | 3 | 5.3 | 110 | 9, 10 |
| Tl | 4 | 4 | 4.8 | 122 | 10 |

field approximation, the interlayer coupling, mediated by a small electron-hopping term, was found to leave $T_c$ practically unaffected. Thus, a three-dimensional pairing mechanism is required to explain the observed dependence of $T_c$ on the average layer spacing.

In this paper we outline our main results and calculate the upper critical field, using the dirty limit of the Ginzburg–Landau (GL) version of our model. Taking the inhomogeneities of intrinsic or extrinsic nature into account, we find corrections to the familiar anisotropic GL behavior, a positive curvature of the upper critical fields, which appears to be a universal feature of layered [12–15] or synthetically layered superconductors.

Our mean-field analysis strongly suggests a weak-coupling pairing mechanism of three-dimensional nature, and the crucial importance of anisotropy and inhomogeneities of intrinsic or extrinsic origin.

## 2. Model

We consider the Hamiltonian [11–17]

$$\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{\text{int}}, \tag{1}$$

where

$$\mathcal{H}_0 = \sum_{\vec{k}_\parallel,\sigma,\ell} [\mathcal{E}_{\vec{k}_\parallel,\ell} C^+_{\vec{k}_\parallel,\sigma,\ell} C_{\vec{k}_\parallel,\sigma,\ell} + t_\ell (C^+_{\vec{k}_\parallel,\sigma,\ell} C_{\vec{k}_\parallel,\sigma,\ell+1} + h.t.)] \tag{2}$$

describes the band structure and

$$\mathcal{H}_{\text{int}} = -\sum_{\vec{k}_\parallel,\vec{k}'_\parallel,\vec{k}''_\parallel,\sigma,\ell} g_\ell(s) C^+_{\vec{k}_\parallel,\sigma,\ell} \cdot C^+_{-\vec{k}_\parallel+\vec{k}''_\parallel,\sigma,\ell} C_{-\vec{k}'_\parallel+\vec{k}''_\parallel,-\sigma,\ell} C_{\vec{k}'_\parallel,\sigma,\ell} \tag{3}$$

the local and attractive pairing interaction, $C^+$ and $C$ denote Fermion creation and annihilation operators, $\ell$ labels the layers, and we assume a periodic sequence of sheets with $q$ layers per period of length $c$. The two-dimensional band structure of the sheets $\mathcal{E}_{\vec{k}_\parallel}$, the coupling between the layers $t$ due to single-particle tunneling, and the coupling constants $g_\ell$ are allowed to vary within a period. The spectrum of $\mathcal{H}$ is labeled by the wavevectors $\vec{k}_\parallel = (k_x, k_y)$ within a layer, $-(\pi/a) \le k_\alpha \le (\pi/a)$, $\alpha = x, y$, and $k_\perp$ is the wavenumber perpendicular to the planes ($z$-direction).

Band structure calculations [18–21], resistivity [22], and upper critical field measurements [14–16] revealed that

$$\frac{W_\perp}{W_\parallel} \ll 1, \quad \frac{\xi^\perp(0)}{\xi^\parallel(0)} \ll 1, \quad \frac{\rho_\perp}{\rho_\parallel} \ll 1, \quad \xi^\perp(0) < s, \tag{4}$$

where $W$ denotes the bandwidth, $\xi(0)$ the zero-temperature correlation length, and $s = c/q$ the mean spacing between the layers. $\perp$ stands for the $z$-direction and $\parallel$ labels a direction within or parallel to the sheets. In **Table 1** we also summarize current experimental data for $T_c$ and the corresponding mean spacing $s$ for the Bi and Tl families, revealing the experimental fact that $T_c$ rises with decreasing mean spacing. Noting that $W_\perp \sim t$ and $\xi^\perp(0) < s$, it is clear that the hopping integrals between the layers are small. Thus, modeling these systems as weakly coupled sheets, or in other words, as layered superconductors, is very appropriate. In this view it is not surprising that the hopping integrals enter $T_c$ as an exponentially small correction only. The observed rise in $T_c$ with decreasing mean spacing $s$ must then be attributed to a three-dimensional pairing mechanism of strength $g$, which obviously depends on $s$. Relating the electron density between the layers to the average spacing $s$ through the relation $\rho \sim s^{-1}$, and assuming strong screening, one finds [11]

$$g(s) \sim s^{-2/3}. \tag{5}$$

## 3. Calculation of the transition temperature

To calculate the transition temperature within the framework of the mean-field approximation, it is sufficient to consider the linearized gap equations [23, 24]

$$\Delta_\ell = \sum_{\ell'} C_{\ell\ell'} \Delta_{\ell'}, \tag{6}$$

where

$$C_{\ell\ell'} = -g_\ell(s)\, k_B T \sum_{\vec{k}_\parallel,\omega_\nu} G^0_{\ell\ell'}(\vec{k}_\parallel, \omega_\nu) G^0_{\ell\ell'}(-\vec{k}_\parallel, -\omega_\nu). \tag{7}$$

Green's function $G^0$ satisfies the equation of motion

$$(i\hbar\omega_\nu - \mathcal{E}_{\vec{k}_\parallel,\ell}) G^0_{\ell\ell'}(\vec{k}_\parallel, \omega_\nu) = \delta_{\ell\ell'} + t_{\ell'} G_{\ell\ell'+1}(\vec{k}_\parallel, \omega_\nu) + t_{\ell'-1} G_{\ell\ell'-1}(\vec{k}_\parallel, \omega_\nu). \tag{8}$$

$T_c$ then follows from the eigenvalue of

$$|C - VE| = 0, \tag{9}$$

yielding, in terms of

$$V_\lambda(k_\perp, T_c) = 1, \qquad \lambda = 1, \cdots, q, \tag{10}$$

the largest $T_c$; $q$ denotes the number of layers per period. In view of the experimental facts [Equation (4)], the solution becomes simplified in terms of

$$k_B T_c \gg W_\perp, \quad \frac{W_\perp}{W_\parallel} \ll 1. \tag{11}$$

Moreover, we assume that the density of states is not singular at the Fermi level, and we adopt a weak-coupling treatment, so that

$$T_c \ll \theta, \tag{12}$$

where $\theta$ is the cutoff temperature. To leading order in the hopping integrals $t$, we then obtain [11, 17]

$$T_c \approx 1.13\theta \exp - \frac{1}{g(s)N_\parallel(0)}. \tag{13}$$

where $g(s)N_\parallel(0)$ refers to the layer within the unit cell yielding the maximum value. $N_\parallel(0)$ is the density of states of that layer. The exponentially small correction term due to interlayer hopping for $q = 1$ is given [25] by

$$T_c(t) = T_c(t = 0) \exp - \frac{t^2}{2\pi k_B T_c(0) W_\parallel}. \tag{14}$$

Thus, the critical temperature depends on the mean spacing only in terms of the pairing coupling constant $g$, but is practically independent [Equation (5)] of the hopping integrals. Nevertheless, the $t_\ell$ must be finite to allow for long-range order.

To compare (13) with the measured $T_c$ and the associated spacing $s$, we rewrite it in the form

$$\ln \frac{T_c}{T_0} = \ln \frac{1.13\theta}{T_0} - \frac{1}{g(s)N_\parallel(0)} \tag{15}$$

Guided by the band structure calculations, we assume $N_\parallel(0)$ to be constant within one family. **Figure 1** shows the experimental results for $\ln T_c/T_0$ versus $s^{2/3}$. They exhibit a clear $s$-dependence, consistent with $s^{2/3}$, expected for strong screening [Equation (5)]. Thus, we are led to the conclusion that the pairing mechanism is of three-dimensional nature. The linear fit through the experimental points, depicted in Figure 1, yields the cutoff temperatures and parameters $g(s)N_\parallel(0)s^{2/3}$ listed in **Table 2**. These parameter values point to a weak-coupling mechanism ($T_c \ll \theta$) with an intermediate boson of rather high energy ($k_B\theta$). Moreover, they justify our weak-coupling assumption [Equation (12)] in solving the gap equation.

## 4. The upper critical field

In the dirty limit the Ginzburg–Landau free-energy functional of our model reads [26]

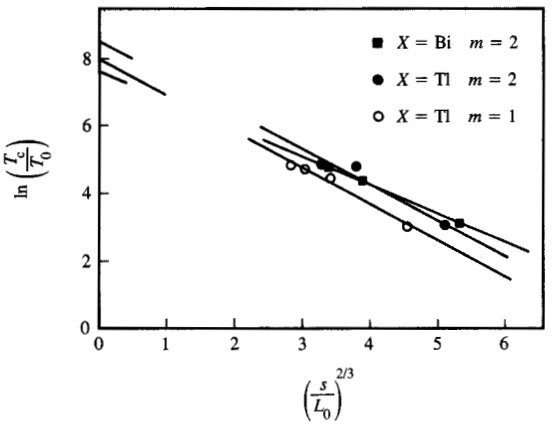


**Figure 1**

Critical temperature $T_c$ versus $s^{2/3}$ for $X_m Ca_{n-1} Ba_2 Cu_n O_{2(n+1)+m}$. $s$ is the mean layer spacing, $T_0 = 1$ K, and $L_0 = 1$ Å.

**Table 2** Estimates of the cutoff temperature $\theta$ and coupling constant $g(s)N_\parallel(0)s^{2/3}$ as obtained from the linear fits shown in Figure 1.

| | $\theta$ (K) | $g(s)N_\parallel(0)s^{2/3}$ |
|---|---|---|
| $Bi_2Ca_{n-1}Ba_2Cu_nO_{2(m+1)+2}$ | 1860 | 1.18 |
| $Tl_2Ca_{n-1}Ba_2Cu_nO_{2(n+1)+2}$ | 4530 | 0.93 |
| $Tl_1Ca_{n-1}Ba\,Cu_nO_{2(n+1)+1}$ | 2660 | 0.93 |

$$F = \int d^2r \sum_\ell \left\{ \frac{\hbar D_\ell}{2} \left[ \left( i\nabla_\parallel + \frac{2e}{\hbar c}\vec{A} \right) \Psi_\ell(\vec{r}) \right]^2 + \frac{m}{M_\ell s_\ell^2} \left| \Psi_{\ell+1}(\vec{r}) \exp i\frac{2\pi}{\theta_0} Hs_\ell x \sin\theta - \Psi_\ell(\vec{r}) \right|^2 + \alpha_\ell |\Psi_\ell(\vec{r})|^2 \right\}. \tag{16}$$

Here $s_\ell$ is the separation of layers $\ell$ and $\ell + 1$, $(\vec{A}, A_z) = (0, x\cos\theta, -\sin\theta)$; $D = 1/2\ V_F^2\tau$ is the two-dimensional diffusion constant; $m$ and $M$ are the effective masses parallel and perpendicular to the layers, respectively. The ratio $m/M$ determines the strength of the interlayer coupling. Finally, $\alpha_\ell \sim \xi_\parallel^{-2}(T)$ in Equation (16) can be derived from the microscopic model [Equations (1)–(3)] in the limit [26]

$$\tau_\ell^2 < \frac{M_\ell s_\ell^2}{m V_{F\ell}^2}, \tag{17}$$

that is, when the scattering time for an electron in a layer is short compared to the time required for an electron to tunnel to the adjacent layer. The functional (16) is an extension of the Lawrence–Doniach expression [27] allowing the properties of the layers and their coupling to vary within a period. If we consider the limit

$$D_\ell \to D, \qquad M_\ell \to M, \qquad s_\ell \to s, \qquad \alpha_\ell \to \alpha, \tag{18}$$

and $s \to 0$, the functional reduces to the familiar form for a dirty and anisotropic superconductor. The upper critical field is then given [16] by

$$H_{c2}^{\perp}(t) = \frac{\phi_0}{2\pi}\frac{1}{\xi^{\parallel}(0)^2}(1-t),$$

$$H_{c2}^{\parallel}(t) = \frac{\phi_0}{2\pi}\frac{1}{\xi^{\parallel}(0)\xi^{\perp}(0)}(1-t), \tag{19}$$

where

$$\frac{\xi^{\perp}(0)}{\xi^{\parallel}(0)} = \sqrt{\frac{m}{M}},$$

$$\frac{1}{\xi^{\parallel}(0)^2} = -\alpha(0)\frac{2}{\hbar D}, \qquad t = \frac{T}{T_c}. \tag{20}$$

The functional (16) is more general, however, taking into account the discrete nature of the interlayer coupling and the variations in the layer properties within a period. In fact, the anisotropic Ginzburg–Landau results apply to layered systems only when the order parameter varies slowly on the scale of the layer separation, which limits their validity to an ever-decreasing range of temperatures near $T$ as the interlayer coupling is decreased.

To explore the variation of layer properties, we next allow small variations in the diffusion coefficient $D$, namely

$$D_\ell = D + \Delta D_\ell, \tag{21}$$

while all other parameters are kept constant. This variation might be of intrinsic nature, due to inequivalent layers within a period or due to inhomogeneities signaled by a broad transition to the superconducting state. Using Equation (16), the evaluation of the upper critical field leads to the eigenvalue problem

$$\left(i\nabla_{\parallel} + \frac{2e}{\hbar c}\vec{A}\right)^2 \Psi_\ell(\vec{r}) - \frac{m}{MS^2}\left(\Psi_{\ell+1}(\vec{r})\exp i\frac{2\pi}{\theta_0}HSx\sin\theta + \Psi_{\ell-1}\exp -i\frac{2\pi}{\theta_0}HSx\sin\theta - 2\Psi_\ell\right) + \frac{2\alpha}{\hbar D_\ell}\Psi_\ell = 0. \tag{22}$$

Assuming small variations in $D_\ell$ only, second-order perturbation theory yields

$$\frac{4\pi H_{c2}^{\perp}}{\phi_0} = \frac{1}{\xi_{\parallel}^2(0)}(1-t)\left\{1 + (1-t)\left(\frac{s}{\xi^{\perp}(0)}\right)^2 F\right\} \tag{23}$$

and

$$\frac{H_{c2}^{\parallel}(t)}{H_{c2}^{\perp}(t)} = \frac{\xi^{\parallel}(0)}{\xi^{\perp}(0)}, \tag{24}$$

where

$$F = \frac{1}{2\pi}\int d(k_\perp s)\frac{|D(k_\perp)|^2}{(k_\perp s)^2 D_0^2}. \tag{25}$$

Thus, $H_{c2}^{\perp}$ and $H_{c2}^{\parallel}$ will exhibit, up to the scaling factor of $\xi^{\parallel}(0)/\xi^{\perp}(0)$, an upward curvature arising from a diffusion constant, varying along the $z$-direction, with an amplitude depending on the ratio squared between the mean spacing and the perpendicular correlation length $L_0$. This ratio is particularly large in the superconductors considered here [Equation (4)], yielding a pronounced positive curvature even for small inhomogeneities ($F \ll 1$), as observed experimentally [13–15].


## Acknowledgments

It is a pleasure to acknowledge stimulating discussions with A. Aharony, D. Baeriswyl, A. Baratoff, K. A. Muller, H. De Raedt, and Z. Tesanovic.


## References


1. J. G. Bednorz and K. A. Mueller, *Z. Phys. B* **64,** 189 (1986).
2. C. Michel, M. Hervieu, M. M. Borel, A. Grandin, F. Deslandes, J. Provost, and B. Raveau, *Z. Phys. B* **68,** 421 (1987).
3. S. Kondoh, Y. Ando, M. Onoda, M. Sato, and J. Akimitsu, *Solid State Commun.* **65,** 1329 (1988).
4. H. Maeda, Y. Tanaka, M. Fukutomi, and T. Asano, *Jpn. J. Appl. Phys. Lett.* **27L,** 209 (1988).
5. Z. Z. Sheng, A. M. Hermann, A. El Ali, C. Almasan, J. Estrada, T. Datta, and R. J. Matson, *Phys. Rev. Lett.* **60,** 937 (1988).
6. R. M. Hazen, L. W. Finger, R. J. Angel, C. T. Prewitt, N. L. Ross, C. G. Hadidiacos, P. J. Heaney, D. R. Veblen, Z. Z. Sheng, A. El Ali, and A. M. Hermann, *Phys. Rev. Lett.* **60,** 1657 (1988).
7. H. V. Zandbergen, Y. K. Huang, M. J. V. Menken, J. N. Li, K. Kadowaki, A. A. Menovsky, G. van Tendeloo, and S. Amelinckx, *Nature* **332,** 620 (1988).
8. S. S. P. Parkin, V. Y. Lee, E. M. Engler, A. I. Nazzal, T. C. Huang, G. Gorman, R. Savoy, and R. Beyers, *Phys. Rev. Lett.* **60,** 2539 (1988).
9. S. S. Parkin, V. J. Lee, A. I. Nazzal, R. Savoy, T. C. Huang, G. Gorman, and R. Beyers, unpublished work.
10. H. Ihara, R. Sugise, M. Hirabayashi, N. Terada, M. Jo, K. Hayashi, A. Negishi, M. Tokumoto, Y. Kimura, and T. Shimomura, *Nature* **334,** 510 (1988).
11. T. Schneider and D. Baeriswyl, *Z. Phys. B* **73,** 5 (1988).
12. J. A. Woollam, R. B. Sowoano, and P. O'Connor, *Phys. Rev. Lett.* **32,** 712 (1974).
13. Y. Koie, T. Nakanomyo, and T. Fukase, *Jpn. J. Appl. Phys.* **27L,** 841 (1988).
14. Y. Hidaka, M. Oda, M. Suzuh, Y. Maeda, Y. Enomoto, and P. Murakawi, *Jpn. J. Appl. Phys.* **27L,** 538 (1988).
15. P. P. M. Palstra, B. Batlogg, L. F. Schneemeyer, and R. J. Cava, preprint.

16. S. T. Ruggiero and M. R. Beasley, in *Synthetic Modulated Structures*, L. Chang and B. C. Giessen, Eds., Academic Press, Inc., New York, 1985, p. 365.
17. D. Baeriswyl and T. Schneider, unpublished work.
18. M. S. Hybertsen and L. F. Mattheiss, *Phys. Rev. Lett.* **60,** 1661 (1988).
19. H. Krakauer and W. E. Pickett, *Phys. Rev. Lett.* **60,** 1665 (1988).
20. R. V. Kasowski, W. Y. Hsu, and F. Herman, submitted to *Phys. Rev. B.*
21. Yu Jaejun, S. Massida, and A. J. Freeman, *Physica C* **152,** 273 (1988).
22. S. Martin, A. T. Fiory, R. M. Fleming, L. S. Schneemeyer, and J. V. Waszcak, *Phys. Rev. Lett.* **60,** 2194 (1988).
23. R. A. Klemm, A. Luther, and M. R. Beasley, *Phys. Rev. B* **12,** 877 (1975).
24. R. A. Klemm and K. Sharnberg, *Phys. Rev. B* **24,** 6361 (1981).
25. Z. Tesanovic, *Phys. Rev. B* **36,** 2364 (1987).
26. R. A. Klemm, M. R. Beasley, and A. Luther, *J. Low Temp. Phys.* **16,** 607 (1974).
27. W. E. Lawrence and S. Doniach, *Proceedings of the 12th International Conference on Low Temperature Physics*, E. Kanda, Ed., Academic Press of Japan, Kyoto, 1971, p. 361.

*Received September 30, 1988; accepted for publication October 3, 1988*

**T. Schneider** *IBM Research Division, Zurich Research Laboratory, Säumerstrasse 4, 8803 Rüschlikon, Switzerland.* After obtaining his diploma at ETH Zurich in 1957, Dr. Schneider spent two years with the Physical Laboratories of the Albis Werke AG and two years with the Physical Laboratory of the Ciba AG, Basel. He then did graduate work at the University of Basel and obtained his Ph.D. in theoretical physics there in 1964. Between 1964 and 1970 Dr. Schneider was in charge of a group of physicists at the Swiss Federal Institute of Reactor Research in Wurenlingen. He joined the Theoretical Physics Group at the IBM Zurich Research Laboratory, Rüschlikon, in January of 1970 and was promoted to the position of Manager, Theoretical Physics, in November of 1970. During 1967 and 1968, Dr. Schneider was a lecturer at ETH. Since 1969 he has been a lecturer at the University of Zurich.