

Thermodynamic and other properties of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$

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We have calculated the thermodynamic, electromagnetic, and critical-field properties of superconducting $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$, using the electron-phonon spectral density determined recently by Weber, from band-structure calculations. Even though the electron-phonon mass enhancement is $\lambda=2.6$, the predicted dimensionless ratios are not very different from those of Pb. For example, the normalized specific-heat jump at T_c , $\Delta C/\gamma(0)T_c=2.8$, the ratio of twice the energy gap to T_c , $2\Delta_0/k_B T_c=5.3$ and $\gamma(0)T_c^2/H_c^2(0)=0.124$ where $H_c(0)$ is the zero-temperature (T) thermodynamic critical field. The reduced upper critical field $h_{c2}(0)$ at $T=0$ in the dirty limit equals 0.76 and shows little Pauli limiting in calculations with $H_{c2}(0)\approx 43$ T.

I. INTRODUCTION

The discovery of superconductivity in the Ba-La-Cu-O system with onset around 30 K, by Bednorz and Müller,¹ has led to a great deal of interest in this family of superconducting oxides and to the observation of superconductivity above 90 K in Y-Ba-Cu-O.² While, so far, much of the activity in this field has been directed towards experimental results, it is very important to try to understand the mechanism responsible for their superconductivity. A particularly suitable material to study in this regard, is $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$, not only because there now exists a considerable amount of experimental data³⁻²¹ on its superconducting properties but, more importantly, because Weber²² has calculated its electron-phonon spectral density $\alpha^2F(\omega)$. His calculations are based on first-principle band-structure calculations and give a spectral density large enough to yield a critical temperature of 35 K for a reasonable value of the Coulomb pseudopotential μ^* . It is important to test the suggestion further, since rival theories have been put forward.²³⁻²⁶

In this paper we calculate, from Weber's²² spectral density, several reduced properties of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ in an isotropic approximation of the Eliashberg equations. These calculations involve no adjustable parameters once μ^* is fixed to get the measured critical temperature. We calculate, as well, several properties which require further input parameters, namely, the electron density of states $N(0)$, and the Fermi velocity v_F . Most of these properties simply scale with either $N(0)$ or v_F^2 [a notable exception being $H_{c2}(T)$ with Pauli limiting, though the dependence is weak]. This enables us to choose values of $N(0)$ and v_F for the purpose of presentation. As experimental determinations or theoretical estimates of these quantities improve in the future, one can easily incorporate the improvements into the results we present without further numerical work. The reduced properties (except for Pauli limiting effects) will remain unchanged. By comparing our theoretical results with experiment it should be possible, in principle, to confirm or rule out the electron-phonon interaction as the mechanism for superconductivity. In practice, some contradictions in the available data, unknown uncertainties in the spectral density, as well as

the neglect of anisotropy, make definitive conclusions difficult and point clearly to a need for more precise experiments.

In Sec. II we calculate thermodynamic properties, and in Sec. III the ratio of the gap to the critical temperature. Section IV deals with the upper critical magnetic field $H_{c2}(T)$ and the Ginzburg-Landau parameter $\kappa_1(T)$, while in Sec. V the electromagnetic coherence length and London penetration depth are calculated. We briefly discuss the normal-state specific heat in Sec. VI. In Sec. VII we draw some conclusions.

II. THERMODYNAMICS

The electron-phonon spectral density for $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ calculated by Weber²² is reproduced in our Fig. 1. With this $\alpha^2F(\omega)$, which corresponds to a mass enhancement parameter $\lambda=2\int_0^\infty[\alpha^2F(\omega)/\omega]d\omega=2.6$, a value of $\mu^*=0.14$ gives the observed critical temperature $T_c=35$ K (Ref. 10) on solution of the linearized Eliashberg equations on the imaginary frequency axis. It will not be necessary to give the finite-temperature Eliashberg equations here because they are well known and standard.²⁷⁻²⁹ They involve gaps $\Delta(i\omega_n)$ and renormalized Matsubara frequencies $\tilde{\omega}(i\omega_n)$ with $i\omega_n=iT_c\pi(2n-1)$, $n=0, \pm 1, \pm 2, \pm 3, \dots$, from which the free-energy difference between the normal and superconducting states (ΔF) follows.²⁸ ΔF is also proportional to the single-spin

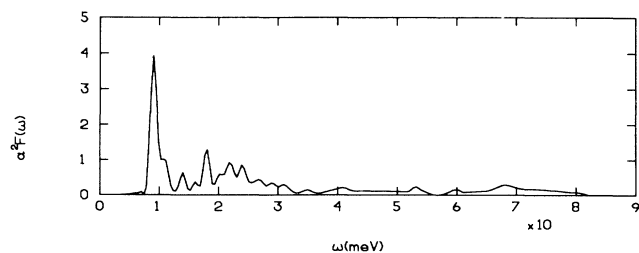


FIG. 1. The electron-phonon spectral density $\alpha^2F(\omega)$ for $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ calculated by Weber (Ref. 22).

electronic density of states at the Fermi energy $N(0)$.

The thermodynamic properties follow directly from the free-energy difference ΔF . Before presenting our results, we need to introduce the normal-state zero-temperature Sommerfeld constant $\gamma(0)$, which will be used for normalization purposes. It is given by

$$\gamma(0) = \frac{2}{3} \pi^2 k_B^2 N(0) (1 + \lambda) . \quad (1)$$

The normalized specific-heat jump at T_c is independent of $N(0)$ or v_F and, hence, depends solely on $\alpha^2 F(\omega)$ and (weakly) on μ^* . We obtain the result $\Delta C(T_c)/\gamma(0)T_c = 2.8$ comparable to the value for Pb and approximately twice the BCS value of 1.43. Comparing this number to experiment is difficult, for two reasons. First, the measurement of $\Delta C(T_c)$ is difficult, because of the large phonon background. The entries in Table I will attest to this fact, as there is considerable disagreement between the first entry and the second two. Second, the value of $\gamma(0)$ is very difficult to extract from experiment. In fact, several groups^{9,20} use the measured value of $\Delta C(T_c)/T_c$ together with the BCS relation $\Delta C(T_c)/\gamma(0)T_c = 1.43$ to extract $\gamma(0)$. We will follow the same procedure using, however, the value 2.8 obtained above instead of 1.43. Furthermore, on the basis of the available data, we choose a value of $\Delta C(T_c)/T_c$ equal to 17 mJ/(mole K²), acknowledging that Batlogg *et al.*⁹

TABLE I. Thermodynamic properties of superconducting states.

Property	Theory	Experiment
$\frac{\Delta C(T_c)}{T_c} \left(\frac{\text{mJ}}{\text{mole K}^2} \right)$	17 ^a	7.6 ± 1.8 ^b 20 ± 5 ^c 16.8 ^d
$\frac{-dH_c(T)}{dT} \Big _{T_c} \left(\frac{\text{mT}}{\text{K}} \right)$	17	14 ^e
$\frac{2\Delta_0/k_B T_c}{2\Delta_0/k_B T_c}$	5.3	2.4 ^f 1.6–2.7 ^g 2.5 ^h 2.9–4.5 ⁱ 5.2–9.1 ^j < 4.5 ^k 5–8.7 ^l
$\Delta C(T_c)/\gamma(0)T_c$	2.8	
$\gamma(0)T_c^2/H_c^2(0)$	0.124	

^aThis value is obtained by demanding $\Delta C(T_c)/T_c$ to be close to the values of c and d. This implies $\gamma(0) \approx 6$ mJ/(mole K).

^bReference 9.

^cReference 16.

^dReference 20.

^eReference 19.

^fReference 5.

^gReference 6.

^hReference 7.

ⁱReference 18.

^jReference 14.

^kReference 17.

^lReference 21.

have measured a value considerably lower. We find that $\gamma(0) = 6.1$ mJ/(mole K²). We emphasize that this value has been obtained from a combination of the (theoretical) result $\Delta C/\gamma T_c = 2.8$ obtained from Weber's²² spectrum, and the (experimental) result that two groups^{16,20} have measured $\Delta C(T_c)/T_c$ to be roughly 17 mJ/(mole K²). Note that our derived $\gamma(0)$ is the same as Batlogg *et al.*⁹ This is entirely fortuitous.

It is now a simple matter to extract the single-spin electron density of states $N(0)$ through the relation (1). We find $N(0) = 0.36$ states/eV Cu atom. This is the value we shall use in the remainder of the paper. Note that it is in sharp disagreement with the theoretical value 0.9 states/eV Cu atom obtained by Freeman, Yu, and Fu.³⁰ This is a greater discrepancy than is found for conventional cases²⁸ and may indicate that Weber's λ is much too large. We will return to this point in Sec. IV when we discuss measurements of the upper critical magnetic field.

The thermodynamic critical magnetic field $H_c(T)$ also follows from the free energy [$H_c^2(t)8\pi = \Delta F$] and is completely determined from $N(0)$ and $\alpha^2 F(\omega)$. It is common practice to present results in terms of reduced ratios, and we follow this convention here. The critical-field deviation function $D(t) \equiv H_c(T)/H_c(0) - (1 - t^2)$ is positive definite at all temperatures with a maximum value of 0.025 at $t = T/T_c = 0.55$, which is similar to the result for Pb. Moreover, the reduced ratio $\gamma(0)T_c^2/H_c^2(0) = 0.124$, is considerably smaller than the BCS value (0.168), indicating once more that BCS relationships should not be used in the analysis of data for this compound because it falls in the strong coupling regime. In making this statement we are assuming, of course, that it is indeed the electron-phonon interaction that is responsible for the superconductivity of this material. This may, in fact, not be the case. These two results are theoretically appealing, in that they are both independent of the density of states, $N(0)$. At the same time, however, there are no direct measurements of $H_c(0)$, so that a comparison with experiment is impossible. Finnemore *et al.*,¹⁹ however, have obtained experimentally a value of $[-dH_c(T)/dT]_{T_c} = 14$ mT/K. This was obtained from measurements of magnetization as a function of applied field for several temperatures, and a correction to account for the granular nature of their sample has been included. Eliashberg theory, with $N(0) = 0.36$ states/eV Cu atom, gives 17 mT/K. The agreement is quite good, and could be improved by a choice of lower $N(0)$ [meaning our theoretical value for $\Delta C(T_c)/T_c$ would be lower].

III. GAP TO CRITICAL TEMPERATURE RATIO

From our solutions of the Eliashberg equations on the imaginary Matsubara frequency axis, we can calculate the gap for real frequencies using an analytic continuation technique.^{31,32} In this way, the gap edge Δ_0 can be determined and the dimensionless ratio $2\Delta_0/k_B T_c$ found to be 5.3, which is close to the value recently suggested in the theoretical work of Marsiglio and Carbotte.³³ Unfortunately, there remains considerable controversy about the experimental value of the gap in La_{1.85}Sr_{0.15}CuO₄.

The tremendous spread in experimental values is obvious from Table I. In general, far-infrared measurements⁵⁻⁷ give a ratio $2\Delta_0/k_B T_c$, which is considerably smaller than the BCS value of 3.54 which is, of course, in disagreement with our theoretical value. On the other hand, one early experiment¹² gave 3.5, which was later reinterpreted,¹⁸ assuming the possibility of gap anisotropy, to yield values from 2.9 to 4.5. It should be noted that all experiments, so far, give considerable absorption below twice the gap edge where there should be none. This absorption, of unknown origin, obscures the identification of the expected rapid rise in absorption as we cross $2\Delta_0$. In contrast to the far-infrared measurements, the early gap values observed for $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ in point contact tunneling experiments have been very large. Hawley, Gray, Capone, and Hinks¹⁴ quote values from 8 to 14 meV. The lower value gives $2\Delta_0/k_B T_c \equiv 5.2$ in good agreement with our theoretical estimate. It should be stressed at this point that our calculations are for an isotropic superconductor. The large anisotropy in the electron-phonon interaction found in the calculations of Weber has been neglected. This anisotropy should lead directly to gap anisotropy in a pure single crystal. For an actual polycrystalline sample, with short mean-free path, much of the anisotropy should have been washed out, however, this may not be very significant effect. Having said this, we note that the more recent experiments by Kirtley *et al.*,¹⁷ using a low-temperature scanning tunneling microscope, give a maximum gap value of 7 meV which corresponds to $2\Delta_0/k_B T_c \sim 4.5$. It is clear that more experiments are needed but that, at present, our theoretical value for $2\Delta_0/k_B T_c$ is not ruled out by experiment.

IV. UPPER CRITICAL MAGNETIC FIELD

We turn now to a discussion of the upper critical magnetic field $H_{c2}(T)$. The strong coupling equations for $H_{c2}(T)$, including Pauli limiting, are a set of two coupled equations of the form^{34,35}

$$\begin{aligned} \tilde{\Delta}(i\omega_n) = \pi T \sum_m [\lambda(m-n) - \mu^*] \hat{\chi}_m \tilde{\Delta}(i\omega_m) \\ + \pi t + \hat{\chi}_n \tilde{\Delta}(i\omega_n), \end{aligned} \quad (2)$$

where $\tilde{\Delta}(i\omega_n)$ is a column vector for the real and imaginary part of the pairing energy

$$\tilde{\Delta}(i\omega_n) = \begin{pmatrix} \tilde{\Delta}_r(i\omega_n) \\ \tilde{\Delta}_i(i\omega_n) \end{pmatrix}, \quad (3)$$

and $\hat{\chi}_m$ is a 2×2 matrix with $\chi_r(\tilde{\omega}_m^0)$ on the diagonal and plus and minus $\chi_i(\tilde{\omega}_n^0)$ on the lower and upper off diagonal respectively, with

$$\chi(\tilde{\omega}_n^0) = \frac{2}{\sqrt{\alpha}} \int_0^\infty dq e^{-q^2} \tan^{-1} \left(\frac{q\sqrt{\alpha}}{|\tilde{\omega}_n^0| + ic\alpha \text{sgn}(\tilde{\omega}_n)} \right); \quad (4)$$

here $c = 1/mv_F^2$, where m is the electron mass, v_F the Fermi velocity, $\alpha = \frac{1}{2} eH_{c2}(T)v_F^2$, and e is the electron charge. Equation (3) applies for any impurity content

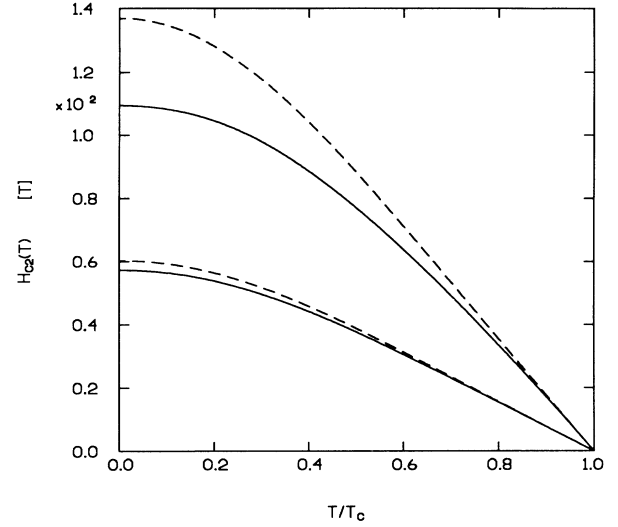


FIG. 2. The upper critical field $H_{c2}(T)$ with (solid line), and without (dashed line), Pauli limiting for two values of the slope, $-[dH_{c2}(T)/dT]_{T_c} = 5$ T/K (top set of curves) and 2.2 T/K (lower set of curves). ($t^+ = 50$ meV.)

$t^+ = 1/2\pi\tau$, with τ the scattering time. Note that $\alpha(T)$ is completely determined from $\alpha^2 F(\omega)$, μ^* , and t^+ . To obtain $H_{c2}(T)$, however, requires a value for v_F^2 . Finally,

$$\lambda(m-n) = 2 \int_0^\infty v \alpha^2 F(v) dv / [v^2 + (\omega_n - \omega_m)^2]. \quad (5)$$

Results for $H_{c2}(T)$ vs T/T_c are given in Fig. 2. The dashed lines were computed without including any Pauli limiting, while the solid curves include this effect. These curves are to be compared with similar ones given in the experimental work of Orlando *et al.*¹¹ The top set was obtained using v_F^2 derived from the case $[-dH_{c2}(T)/dT]_{T_c} = 5.0$ T/K while the lower set are for a slope of 2.2 T/K. Without Pauli limiting, $H_{c2}(0) = 137$ T, which is reduced to 110 T when it is included. This is not very different from the results suggested by Orlando *et al.*,¹¹ in

TABLE II. Theoretical and experimental values of various properties.

Property	Theory		Experiment
	Clean	Dirty	
$H_{c2}(T_c)$ (T/K)			5.0 ^a 2.2 ^b 1.51 ^c
$h_{c2}(0)$ (no Pauli limiting)	0.87	0.78	...
$h_{c2}(0)$ (Pauli limiting)	0.84	0.76	...
Deduced values of	0.17	0.4 ^a	...
v_F ($\times 10^6$ m/s)	0.26	0.6 ^b	...
	0.31	0.73	...
$\frac{\kappa(0)}{\kappa(T_c)}$ (Pauli limiting)	1.75	1.58	...

^aReference 11, using onset data.

^bReference 11, using midpoint data.

^cReference 10.

the case $\lambda=2$, and no spin-orbit scattering. To get sensible results from a BCS analysis, it is clearly necessary to include a factor of λ of the order of 2. The situation is similar when the slope is taken to be 2.2 T/K, in this case, band splitting effects are much smaller and $H_{c2}(0)=57$ with Pauli limiting in the dirty limit. To understand the effect of impurities, we have also done calculations in the clean limit. Our results are perhaps best presented in terms of the reduced critical field $h_{c2}(t)$ given by

$$h_{c2}(t) = \frac{H_{c2}(T)}{T_c |dH_{c2}/dT|_{T_c}}, \quad (6)$$

with $t \equiv T/T_c$.

In the case of no Pauli limiting, this quantity is independent of choice of Fermi velocity and is completely determined from Weber's spectrum.²² With Pauli limiting, we have used a Fermi velocity determined from the measurement of $(dH_{c2}/dT)_{T_c} = -1.51$ T/K by Kwok *et al.*¹⁰ The results for $h_{c2}(0)$ are shown in Table II.

Also shown in Table II are results for values of the Fermi velocity deduced from H_{c2} slope measurements. The results can vary by almost an order of magnitude, depending on the measurement and the analysis. Note that

$$\xi(T) = \frac{hv_F}{2} \left[\frac{\sum_{n=1}^{\infty} \frac{\tilde{\Delta}^2(i\omega_n)}{[\tilde{\omega}^2(i\omega_n) + \tilde{\Delta}^2(i\omega_n)]^{3/2}}}{\sum_{n=1}^{\infty} \frac{\tilde{\Delta}(i\omega_n)}{\tilde{\omega}^2(i\omega_n) + \tilde{\Delta}^2(i\omega_n)}} \right]. \quad (7)$$

This expression is easily evaluated from our finite temperature Eliashberg equation solutions. In the clean limit, we get a value $\xi(0) = 35 \times 10^{-8}$ cm. It is often the case that the BCS version of Eq. (7), namely $\xi^{\text{BCS}}(0) = v_F[\pi\Delta_0(1+\lambda)]$, is used to estimate values for the coherence length. Using this expression, we obtain a value $\xi^{\text{BCS}}(0) = 23 \times 10^{-8}$ cm, which is very different from the more accurate result [$\xi^{\text{BCS}}(0)/\xi(0) = 0.65$]. These results can vary significantly due to uncertainties in v_F . It is

$$\lambda_L(T) = \left[\frac{3c^2}{8\pi N(0)e^2 v_F^2} \right]^{1/2} \left[\sum_{n=1}^{\infty} 2\pi T \frac{\tilde{\Delta}^2(i\omega_n)}{[\tilde{\Delta}^2(i\omega_n) + \tilde{\omega}^2(i\omega_n)]^{3/2}} \right]^{-1/2}. \quad (8)$$

In a BCS-like approximation, the last bracket in Eq. (8) becomes $(1+\lambda)^{1/2}$. In Fig. 3 we show the temperature variation of the ratio $\{[\lambda_L(0)]/[\lambda_L(T)]\}^2 - (1-t^4)$ obtained for $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ (solid line) and compare with Pb (dashed line). This plot indicates that the oxide superconductor is only slightly more strong coupling than is Pb. The zero temperature value $\lambda_L(0)$, using the Fermi velocity $v_F = 0.31 \times 10^8$ cm/sec, is 2600 Å, only slightly different from the BCS estimate corrected for the factor of $(1+\lambda)$, which is 2700 Å. These estimates are somewhat smaller than the value of 3300 Å given in the experimental work of Finnemore *et al.*¹⁹

VI. NORMAL-STATE SPECIFIC HEAT

We briefly discuss here the normal-state electronic specific heat. The electron-phonon interaction renormal-

different values are deduced in Ref. 10, based on a BCS-type analysis. Much of the discrepancy is removed if simple renormalization factors $(1+\lambda)$ are included. For example, one should interpret their value as a renormalized Fermi velocity $v_F^* = v_F/(1+\lambda)$.

Another quantity of interest is the Ginzburg-Landau parameter $\kappa(T)$. It is given by $\kappa = 1/\sqrt{2}H_{c2}(T)/H_c(T)$. Using $|dH_{c2}(T)/dT|_{T_c} = 1.51$, we obtain $\kappa(T_c) = 63$. Finnemore *et al.*¹⁹ deduced a value $\kappa(T_c) = 90$. Both these numbers are essentially experimental, and so indicate the discrepancies that exist in the literature. Note, however, that we determine the ratio $\kappa(0)/\kappa(T_c)$ directly from $\alpha^2 F(\omega)$. As seen in Table II, there is a considerable enhancement above the BCS values of 1.26 (clean limit) and 1.2 (dirty limit).

V. COHERENCE LENGTH AND PENETRATION DEPTH

In a full strong coupling formulation for the electromagnetic properties of a superconductor, the coherence length $\xi(T)$ is given by³⁶

clear, nonetheless, that a complete theory, such as Eq. (7), is needed to get a reliable quantitative value for the coherence length. We also quote, in the clean limit the reduced quantity $\xi(T_c)/\xi(0) = 0.83$, which depends only on $\alpha^2 F(\omega)$. This is not very different from Pb and in fact is not much different from the BCS value of 0.75.

Next we consider the London-limit penetration depth $\lambda_L(T)$ given by³⁶

izes the Sommerfeld γ so that $C_N(T) = \gamma(T)T$, where

$$\gamma(T) = \gamma(0) \left[\frac{1+\lambda(T)}{1+\lambda(0)} \right], \quad (9)$$

where $\lambda(T)/\lambda(0)$ is given by Grimvall,^{37,38} and Kresin and Baitsev,³⁹ and need not be repeated here. Results for $\lambda(T)/\lambda(0)$ are shown in Fig. 4. Because of the large value of the critical temperature and the modest value (in energy) of the important phonon modes in $\alpha^2 F(\omega)$, we find that at $T=T_c$ the renormalized $\gamma(T_c)$ is very different from $\gamma(0)$. This situation is quite different from the conventional case where T_c is relatively small (A15, transition metals, etc.) and the difference between $\gamma(0)$ and $\gamma(T_c)$ is small. This should be kept in mind when analyzing data on the specific heat. If $\gamma(T_c)$ has been used, instead of $\gamma(0)$, to normalize the two dimensionless ratios $[\Delta C(T_c)]/[\gamma(T_c)T_c]$ and $[\gamma(T_c)T_c^2]/[H_c^2(0)]$, we would

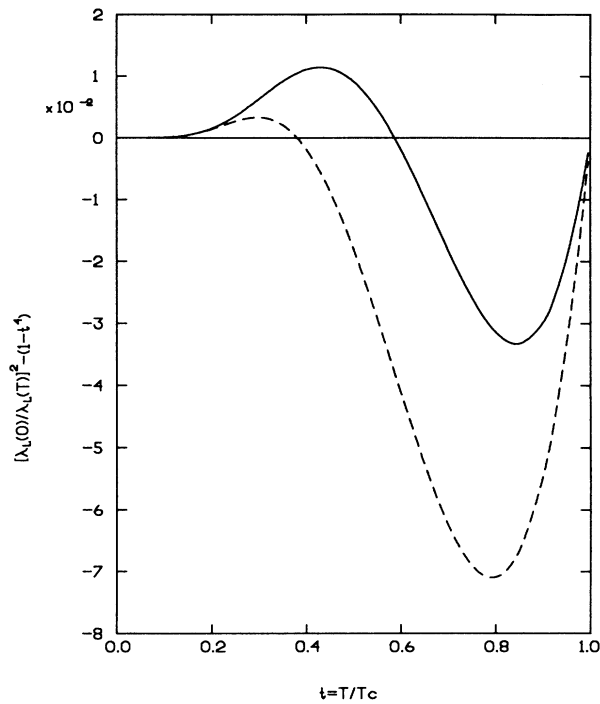


FIG. 3. The temperature variation of the London penetration depth for $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ (solid line) compared with that for Pb (dashed line).

have obtained the values 4.7 and 0.074, respectively, instead of 2.82 and 0.124. It should be pointed out, however, that $\gamma(T_c)$ would be very hard to measure, even in principle, because the electronic specific heat is superimposed on, and strongly coupled to a large phonon background. Also, it is $\gamma(0)$ and not $\gamma(T_c)$ that is derived from the slope of the upper critical magnetic field $H_{c2}(T)$ at T_c .

VII. CONCLUSIONS

In conclusion, we have calculated, from Weber's electron-phonon spectral density, the superconducting properties of $\text{La}_{1.85}\text{Sr}_{0.15}\text{CuO}_4$ and have found that this superconductor is in the strong coupling regime with thermodynamic indices and other such quantities not so different from those for Pb (the classic strong coupler). Whenever possible, we have compared with experiment. Because a considerable amount of uncertainty in the data and its in-

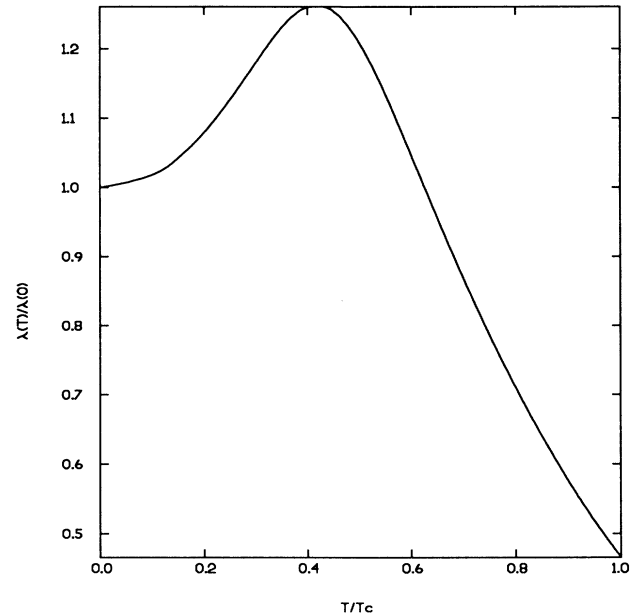


FIG. 4. The Grimvall renormalization factor $\lambda(T)/\lambda(0)$ as a function of reduced temperature $t=T/T_c$ calculated from Weber's spectral density. The value of $\lambda(T)$ at T_c less than half its zero temperature value.

terpretation exists, we are not able, at this point, to use the data to confirm or reject the electron-phonon mechanism. On the other hand, we have required a rather low electronic density of states at the Fermi surface $N(0)$ to accommodate the large λ calculated from Weber's spectrum. The value obtained from theory is more than a factor of 2 larger. Such a large discrepancy is not observed in conventional cases and may indicate a breakdown of the phonon mechanism. We hope that our calculations may stimulate more experiments so as to pin down more accurately the various parameters and, hopefully, allow to a firmer conclusion to be reached. If, for example, some consensus could be reached as to the experimental value of $2\Delta_0/k_B T_c$ or some other property calculated here a more definite conclusion would then be possible.

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