Resistivity of $La_{1.825}Sr_{0.175}CuO_4$ and $YBa_2Cu_3O_7$ to 1100 K: Absence of Saturation and Its Implications

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Resistivity was measured from T_c to 1100 K for $La_{1.825}Sr_{0.175}CuO_4$, where it is linear to 1100 K, and for $YBa_2Cu_3O_7$, where it is linear to 600 K, increases faster than linear at $T > 600$ K, because of loss of oxygen, and changes slope at the 950-K structural transition. The observed absence of saturation in the linear parts of the data implies weak electron-phonon coupling in both superconductors, thus excluding the possibility of phonon-mediated superconductivity. Values of the plasma energy are derived in combination with other experimental data.

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In this Letter we present first measurements of the normal-state resistivity $\rho(T)$ of the high-T_c oxide superconductors[1] from T_c to 1100 K. Aiming to elucidate the strength of electron-phonon interaction and being motivated by the peculiar nearly linear temperature dependence of the resistivity below room temperature reported for $La_{2-x}Sr_xCuO_4$ (LSCO)[2] and $YBa_2Cu_3O_7$ (YBCO),[3] we measured $\rho(T)$ to the highest possible temperatures. Resistivities are essentially linear to 1100 K in LSCO and to 600 K in YBCO. In this work we emphasize the important implication, arguing that resistivity which is linear to high temperature can be understood only in terms of weakly coupled electrons and phonons; if $\rho(T)$ were to have a nonphonon origin, then the phonon coupling would be weaker yet. From our analysis, we derive an *upper-bound* estimate for the strength of the electron-phonon coupling λ, and conclude that high T_c's must be nonphonon in nature for both compounds. It is important to recognize that this conclusion is reached on the basis of linearity of $\rho(T)$ alone, since the less reliable absolute values of resistivity do not figure into the argument. The plasma energy $\hbar\omega_p$ is estimated both from resistivity and from independent measurements of the London penetration depth $\lambda_L(0)$.

Measurements were taken on bar-shaped samples, $\simeq 1\text{-mm}^2$ cross section, that were cut from fully fired ceramic pellets and reannealed in oxygen. Four contact wires, gold or silver, were attached with silver cement. The resistivity data, presented in Fig. 1, are composites of separate cryostat and O_2-furnace runs. The sharp upturn of $\rho(T)$ in $YBa_2Cu_3O_{7-\delta}$ above 600 K can be ascribed to the temperature dependence of the oxygen deficiency[4] $\delta(T)$ which plays the dual role of reducing the number of carriers and increasing the number of scattering centers.[5] By contrast, LSCO is stable in O_2 to 1100 K.[6] Furthermore, in YBCO we observe an abrupt change in slope of $\rho(T)$ at the orthorhombic-to-tetragonal transition at 950 K, as shown in Fig. 2. This demonstrates a potential of resistive measurements to detect the temperature of this transition precisely and to study its behavior in various atmospheres.[5]

As can be seen from Figs. 1 and 2, the slope $\alpha = [\rho(T) - \rho(0)]/T$ of the linear parts of the curves (the region $T < T_2$, with $T_2 = 1100$ K for LSCO and 600 K for YBCO) shows no indication of decreasing at high temperature. Qualitatively, this reveals at once that mean free paths l are much longer than interatomic spacings a at temperatures below T_2. In contrast, $\rho(T)$ in metals with $l \simeq a$ always displays strong deviation from linearity at high temperature, known as resistivity saturation. We have included in Fig. 1 data for V_3Si[7] as an example of such saturation behavior and for Cu as typical linear-metallic resistivity.

Turning now to the absolute values of $\rho(T)$, we note that ceramic samples having $\gtrsim 90\%$ of theoretical density, highest T_c's, and sharp transitions all have similar

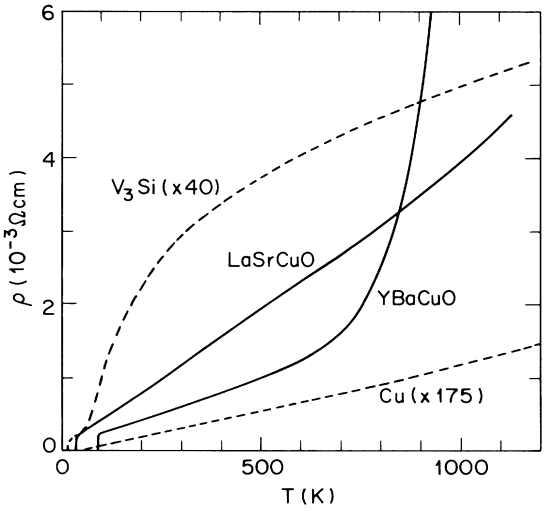

FIG. 1. Temperature dependence of the resistivity for $La_{1.825}Sr_{0.175}CuO_4$ and $YBa_2Cu_3O_{7-\delta}$ measured in this work. Data for V_3Si and Cu are shown for comparison.

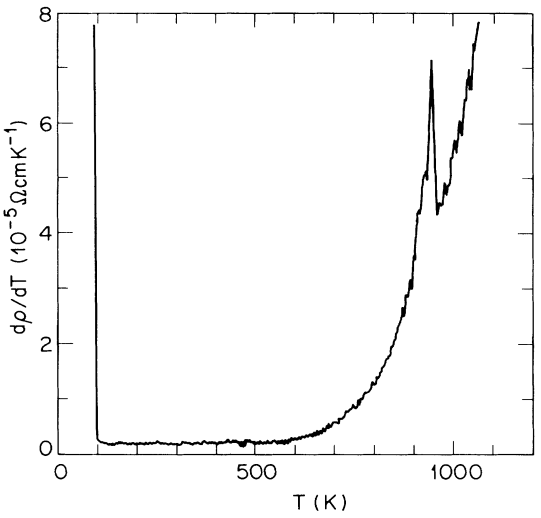

FIG. 2. Derivative of resistivity for $YBa_2Cu_3O_{7-\delta}$, $d\rho/dT \simeq [\rho(T)-\rho(0)]/T$ between 100 and 600 K, showing the break at the orthorhombic-tetragonal structural transition in O_2.

values of $\rho(300\text{ K})$ and α. Our values at 300 K are similar to the ones published by Cava and co-workers.[2,3] Recent work of Enomoto *et al.*[8] on epitaxial films of YBCO shows that the resistivity anisotropy $\rho_\perp/\rho_\parallel$ is at least an order of magnitude, because of preferential conduction within the Cu-O planes. An effective-medium model ($\rho_\parallel \ll \rho_\perp$) of a polycrystalline ceramic leads to a correction factor $\beta=\rho/\rho_\parallel=2$.[9] Comparing data for best ceramics with best epitaxial films grown on $SrTiO_3$[10,11] indicates that $\beta \simeq 2$ is probably the correct factor. As an example, Suzuki and Murakami[10] report for an in-plane $La_{1.94}Sr_{0.06}CuO_4$ film $\rho_\parallel=700\ \mu\Omega$ cm and $\alpha_\parallel=1.9\ \mu\Omega$ cm K^{-1} at 300 K, about a factor of 2 lower than for ceramics. The values for $\rho_\parallel$ and $\alpha_\parallel$ given in Table I were computed with $\beta=2.2$ to account for 90% density and for this anisotropy. If the true intrinsic resistivities turn out to be smaller than stated, this would lower our estimate of $\hbar\omega_p$ from resistivity, but would have no effect on the argument concerning λ, which derives from linearity alone.

We now proceed to combine resistivity results with band-theory parameters in order to calculate l and λ.

TABLE I. Basal-plane intrinsic resistivities ($\rho_\parallel=\rho_{meas}/2.2$), maximum temperature of linearity T_2, and resistivity slope $\alpha_\parallel=[\rho(T_2)-\rho_\parallel(0)]/T_2$.

	$\rho_\parallel(0)$ ($\mu\Omega$ cm)	$\rho_\parallel(300\text{ K})$ ($\mu\Omega$ cm)	$\rho_\parallel(T_2)$ ($\mu\Omega$ cm)	T_2 (K)	$\alpha_\parallel$ ($\mu\Omega$ cm K^{-1})
LSCO	33	534	1980	1100	1.77
YBCO	33	280	555	600	0.87

Expressing total resistivity in terms of the plasma energy, $\rho(T)=4\pi\omega_p^{-2}\tau^{-1}=4\pi v_F\omega_p^{-2}l^{-1}$, we write for the phonon part $\rho(T)=\rho(0)=4\pi\omega_p^{-2}\tau_{e\text{-}ph}^{-1}$ where v_F is the Fermi velocity and $\tau_{e\text{-}ph}^{-1}$ is the electron-phonon scattering rate. It can be expanded at $T \gtrsim \theta_D$ as $\tau_{e\text{-}ph}^{-1}=2\pi\hbar^{-1}\lambda_{tr}kT$, where λ_{tr} is closely related to the McMillan coupling constant λ appearing in the theory of superconductivity.[12,13] We then find

$$pl=4\pi v_F(\omega_p)^{-2}=4.95\times10^{-4}v_F(\hbar\omega_p)^{-2}, \tag{1a}$$

$$\lambda=\frac{\hbar\omega_p^2}{8\pi^2k_B}\frac{\rho(T)-\rho(0)}{T}=0.246(\hbar\omega_p)^2\alpha, \tag{1b}$$

in units $[\alpha]=\mu\Omega$ cm K^{-1}, $[l]=$Å, $[v_F]=$cm s^{-1} and $[\hbar\omega_p]=$eV. Considerable previous experience in the use of these relations to estimate coupling in simple and transition metals[13] and, with additional corrections, $A15$'s[14] had led to the conclusion that band-structure calculations coupled with resistivity correctly predict the strength of electron-phonon coupling in those materials.

Before attempting to couple Eqs. (1) with experimental quantities, we point out that for LSCO Allen, Pickett, and Krakauer[15] recently presented their calculated basal-plane values, $(v_F)_\parallel=2.2\times10^7$ cm s^{-1} and $(\hbar\omega_p)_\parallel=2.9$ eV for [Sr] $x=0.05$, and, with the resistivity data of Ref. 10, calculated $\lambda=3.2$. However, one arrives at large λ at the expense of unphysically small values of l—in our case we find for LSCO $\lambda=3.7$ and $l(T_2)=0.6$ Å. For YBCO we similarly find, using preliminary band-structure results,[16] $(\hbar\omega_p)_\parallel=3.6$ eV and $(v_F)_\parallel=2.5\times10^7$ cm s^{-1}, that $\lambda=2.8$ and $l(T_2)=1.7$ Å. Thus results from band-structure calculations presently contradict the obvious implication of Fig. 1 that $l(T_2)\gg a$, and violate the Mott-Ioffe-Regel (MIR) rule[17]: $l>a$, where a is a minimum scattering length (here taken to be 2 Å, the Cu-O bond length).

We resolve this dilemma by turning to independent experimental evidence for considerably smaller $(\hbar\omega_p)_\parallel$, recognizing that $[\lambda_L(0)]_\parallel=c(1+\lambda)^{1/2}(\omega_p)_\parallel^{-1}$ and explicitly including mass renormalization at low temperature. Experimentally, $\lambda_L(0)$ has been determined by muon-spin-rotation linewidths, $\Lambda_{\mu sr}$, in a technique insensitive to sample surfaces. From recent results, $\Lambda_{\mu sr}=0.9\ \mu s^{-1}$ in LSCO[18] and $3\ \mu s^{-1}$ in YBCO,[19] and accounting for anisotropy,[20] we obtain $[\lambda_L(0)]_\parallel=2200$ Å for LSCO and 1200 Å for YBCO. When we anticipate our estimates for λ, this leads to the $(\hbar\omega_p)_\parallel$ values in Table II. Since in-plane Fermi velocities $(v_F)_\parallel$ are also needed, we deduce them by combining Sommerfeld constant $\gamma=\frac{1}{3}\pi^2k_B^2N_F(0)(1+\lambda)^{1/2}$ with the plasma frequency, written in 2D as $(\omega_p)_\parallel^2=2\pi e^2N_F(0)(v_F)_\parallel^2$, so that

$$(v_F)_\parallel=(\pi/6\gamma)^{1/2}ck_Be^{-1}(1+\lambda)[\lambda_L(0)]_\parallel^{-1}.$$

TABLE II. Estimates of $b=l(T_2)/a, l$ at 300 K, electron-phonon coupling, Fermi velocity, and plasma frequency (from resistivity and from muon spin rotation).

	$b(T_2)$	l(300 K) (Å)	λ	$(v_F)_\parallel$ (10^7 cm s^{-1})	$(\hbar\omega_p)_\parallel^{[\rho]}$ (eV)	$(\hbar\omega_p)_\parallel^{[\mu sr]}$ (eV)
LSCO	7.6	56	0.1	0.95	0.4	0.85
YBCO	5.5	22	0.3	1.4	1.1	1.7

From experimental values of $\gamma=7$ mJ mole^{-1} K$^{-2}=1.1\times10^3$ erg cm^{-3} K^{-2} in LSCO[21] and 9 mJ mole^{-1} K^{-2} (per Cu atom) $=2.3\times10^3$ erg cm^{-3} K^{-2} in YBCO,[22] we obtain $(v_F)_\parallel=0.85(1+\lambda)\times10^7$ cm s^{-1} and $1.08(1+\lambda)\times10^7$ cm s^{-1}, respectively.

Returning to the main focus of this Letter, we demonstrate that long mean free paths at high temperatures exclude strong coupling. If mean free path is at least $b(T)$ times larger than a, $l\geq b(T)a$, then at the highest temperature of linearity T_2 we obtain from Eqs. (1) in 2D

$$\lambda\leq 1.22\times10^{-4}(v_F)_\parallel/b(T_2)aT_2. \tag{2}$$

With ρ and ω_p dropping out, this reveals that linear resistivity at high T_2 by itself restricts λ, in virtue of the imposition $b\gg1$. Indeed, in strongly saturated $A15$'s, such as V_3Si, at 300 K, $b\simeq2$–4.[14] In Nb at 300 K, saturation is hardly visible, and $b\simeq9$.[23] In Cu, where saturation is not seen at all, $b\simeq200$.[23] Following these examples, we take $b(T_2)\geq5$ as a preliminary guess for the oxide superconductors at T_2. Consequently, with our self-consistent values for $(v_F)_\parallel$ and $a=2$ Å, we find from Eq. (2) small coupling constants indeed—$\lambda_{LSCO}\leq0.1$ and $\lambda_{YBCO}\leq0.3$. Furthermore, from Eq. (1a) we obtain

$$(\hbar\omega_p)_\parallel\leq2.22\times10^{-2}[(v_F)_\parallel/\rho_\parallel b(T_2)a]^{1/2}. \tag{3}$$

With $b(T_2)\geq5$, this gives $(\hbar\omega_p)_\parallel^{LSCO}\leq0.5$ eV and $(\hbar\omega_p)_\parallel^{YBCO}\leq1.1$ eV.

The general argument presented above does not depend on a specific model for resistivity saturation. Now, to improve on these estimates, we introduce a specific saturation model, noting first that Eq. (1b) relates λ to not the measured slope α, but rather to an "ideal" slope α_{id} which has been *corrected* for saturation.[14] The correction $C_s=\alpha_{id}/\alpha$ can be obtained from the parallel-resistor model of resistivity saturation,[24] where measured conductivity $\rho^{-1}(T)$ is an "ideal" conductivity $\rho_{id}^{-1}(T)$ in parallel with ρ_s^{-1}, the limiting conductivity at the MIR limit $l=a$. From the model we find[14]

$$C_s=[1-\rho(0)/\rho_s]^{-1}[1-\rho(T)/\rho_s]^{-1}. \tag{4}$$

Rewriting Eq. (1b) as $\lambda=0.246(\hbar\omega_p)_\parallel^2C_s\alpha_\parallel$, and noting that $\rho(0)\ll[\rho_s,\rho(T_2)]$, we recover Eqs. (2) and (3), with b replaced by $b-1$. Noting that we no longer require linearity of $\rho(T)$, we now use Eq. (4) to obtain better quantitative estimates of $b(T_2)$. If saturation were the only source of nonlinearity in $\rho(T)$, then $\alpha_{id}(T)=$const, and from Eq. (4)

$$b=\frac{\rho_s}{\rho(T_2)}=\frac{1-\epsilon_s\rho(T_1)/\rho(T_2)}{1-\epsilon_s}, \tag{5}$$

where $\epsilon_s=\alpha(T_2)/\alpha(T_1)$. The model thus shows explicitly that absolute resistivities do not enter into estimates of b nor, therefore, λ.

Taking $T_1=300$ K we find $\rho(T_1)/\rho(T_2)$ is 0.27 for LSCO and 0.50 for YBCO. The systematic error in α may be $\simeq10\%$; hence, we take $\epsilon_s\geq0.9$, and compute from Eq. (5): $b_{LSCO}\geq7.6$ and $b_{YBCO}\geq5.5$. Considering that we now use $b-1$ in Eqs. (2) and (3), our first guess of $b\geq5$ was not far off. The resulting estimates for $b(T_2)$, l(300 K), λ, $(v_F)_\parallel$, and $(\hbar\omega_p)_\parallel$, are given in Table II. As a check, we use Eqs. (2) and (5) to calculate upper-bound λ estimates of 2.4 for V_3Si, which is most reasonable, and 0.6 for Cu, which is high, reflecting our conservative estimate of linearity.

The next level of sophistication is to account for other sources of nonlinearity, such as the effects of phonon anharmonicity and band-structure broadening. The first, at least as it relates to thermal expansion, is readily calculated[20] and we estimate that between $T_1=300$ K and T_2 it opposes saturation and leads to an increase in slope $\epsilon_{anh}=\alpha(T_2)/\alpha(T_1)$ of 1.1 and 1.04 for the two compounds. The second effect is sensitive to band-structure details, presently under debate.[16] Our computations, based on preliminary results[16,25] indicate that lifetime broadening acts in the same direction as saturation, i.e., decreasing the slopes with increasing T.[26] Considering that anharmonicity and broadening effects oppose each other, and the difficulty of reliable calculation of the latter, we are inclined to omit these corrections in our estimates. It seems highly improbable to us that the observed linearity in $\rho(T)$ arises from accidental precise cancellation between strong saturation and effects opposing saturation, also including the possibility of incipient oxygen desorption.

In conclusion, the observed linear $\rho(T)$ behavior in LSCO (to 1100 K) and YBCO (to 600 K) implies weak electron-phonon coupling in both compounds. Our upper estimates based on linearity are $\lambda_{LSCO}=0.1$ and $\lambda_{YBCO}=0.3$. These estimates could be increased somewhat if strong effects opposing saturation were at work, but we take this to be unlikely. In the McMillan formula for T_c, these values of λ would predict $T_c\simeq0$. Hence, high-T_c superconductivity must be nonphonon in origin. Our upper estimates of $(\hbar\omega_p)_\parallel$, 0.4 eV for LSCO and 1.1 eV for YBCO, are based both on linearity of $\rho(T)$ and on absolute values of $\rho_\parallel$. They are smaller, although not drastically so, than values of 0.85 and 1.7 eV we deduced from penetration-depth measurements, and significantly smaller than band-structure values. Better agreement may be in store if intrinsic $\rho_\parallel$ is found smaller, or penetration depths larger, or both. The dramatic increase in $\rho(T)$ of YBCO above 600 K, related to oxy-

gen desorption, and the break in slope at the structural transition, are examined in detail elsewhere.[5,20]

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