Are high-temperature superconductors in the dirty limit?

C. C. Homes,* S. V. Dordevic, T. Valla, and M. Strongin
Department of Physics, Brookhaven National Laboratory, Upton, New York 11973

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A scaling relation $\rho_s \propto \sigma_{dc} T_c$ has been observed in the copper-oxide superconductors, where ρ_s is the strength of the superconducting condensate, T_c is the critical temperature, and σ_{dc} is the normal-state dc conductivity close to T_c. This scaling relation is examined within the context of a clean and dirty-limit BCS superconductor. These limits are well established for an isotropic BCS gap 2Δ and a normal-state scattering rate $1/\tau$; in the clean limit $1/\tau \ll 2\Delta$, and in the dirty limit $1/\tau > 2\Delta$. The dirty limit may also be defined operationally as the regime where ρ_s varies with $1/\tau$. It is shown that the scaling relation $\rho_s \propto \sigma_{dc} T_c$ is the hallmark of a BCS system in the dirty-limit. While the gap in the copper-oxide superconductors is considered to be *d*-wave with nodes and a gap maximum Δ_0, if $1/\tau > 2\Delta_0$ then the dirty-limit case is preserved. The scaling relation implies that the copper-oxide superconductors are likely to be in the dirty limit, and that as a result the energy scale associated with the formation of the condensate is scaling linearly with T_c.

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Introduction. Scaling laws express a systematic and universal simplicity among complex systems in nature. For example, such laws are of enormous significance in biology, where the scaling relation between body mass and metabolic rate spans 21 orders of magnitude.[1,2] Scaling relations are equally important in the physical sciences. Since the discovery of superconductivity at elevated temperatures in copper-oxide materials[3] there has been considerable effort to find trends and correlations between the physical quantities, as a clue to the origin of the superconductivity.[4] One of the earliest patterns that emerged was the linear scaling of the superfluid density ρ_s ($\propto 1/\lambda^2$, where λ is the superconducting penetration depth) in the copper-oxygen planes of the hole-doped materials with the superconducting transition temperature T_c. This is referred to as the Uemura relation,[5,6] and it works reasonably well for the underdoped materials. However, it does not describe very underdoped,[7] optimally doped (i.e., T_c is a maximum), overdoped,[8,9] or electron-doped materials.[10] A similar attempt to scale ρ_s with the dc conductivity σ_{dc} was only partially successful.[11] We have recently demonstrated that the scaling relation $\rho_s \propto \sigma_{dc} T_c$ may be applied to a large number of high-temperature superconductors, regardless of doping level or type, nature of disorder, crystal structure, or direction (parallel or perpendicular to the copper-oxygen planes).[12] The optical values of $\rho_s(T \ll T_c)$ and $\sigma_{dc}(T \gtrsim T_c)$ have been determined for a large number of copper-oxide superconductors, as well as the bismuth-oxide material $Ba_{1-x}K_xBiO_3$; the results are shown as a log-log plot in Fig. 1 and indicate that within error the points may be described by the relation $\rho_s \propto 35\,\sigma_{dc} T_c$. In addition, the elemental BCS superconductors Nb and Pb (without any special regards to preparation) are also observed to follow this scaling relation reasonably well.

The values for σ_{dc} and ρ_s have been obtained almost exclusively from reflectance measurements from which the complex optical properties have been determined. The dc conductivity has been extrapolated from the real part of the optical conductivity $\sigma_{dc} = \sigma_1(\omega \to 0)$ at $T \gtrsim T_c$. For $T \ll T_c$, the response of the dielectric function to the formation of a condensate is expressed purely by the real part of the dielectric function $\epsilon_1(\omega) = \epsilon_\infty - \omega_{ps}^2/\omega^2$, which allows the strength of the condensate to be calculated from $\omega_{ps}^2 = -\omega^2\epsilon_1(\omega)$ in the $\omega \to 0$ limit. Here, $\omega_{ps}^2 = 4\pi n_s e^2/m^*$ is the square of the superconducting plasma frequency and $\rho_s \equiv \omega_{ps}^2$. The strength of the condensate may also be estimated by tracking the changes in the spectral weight above and below T_c, where the spectral weight is defined as[13] $N(\omega, T) = (120/\pi)\int_{0^+}^{\omega} \sigma_1(\omega', T)\, d\omega'$. The condensate may be calculated from the shift in the spectral weight $\rho_s = N_n - N_s$, where $N_n = N(\omega, T \simeq T_c)$, and $N_s = N_s(\omega, T \ll T_c)$. This is the Ferrell-Glover-Tinkham sum rule which tracks changes in the optical conductivity $\sigma_1(\omega)$ above and below T_c due to the formation of a condensate at zero frequency.[14,15]

A deeper understanding of the scaling relation as it relates to both the elemental superconductors and the copper-oxide materials may be obtained from an examination of the spectral weight above and below T_c in relation to the normal-state scattering rate. When Nb is in the dirty limit, it follows the $\rho_s \propto \sigma_{dc} T_c$ relation, but in the clean limit there is a deviation from this linear behavior. (This result will be explored in more detail shortly.) The terms "clean" and "dirty" originate from the comparison of the isotropic BCS energy gap 2Δ with the normal-state scattering rate $1/\tau$; the clean limit is taken as $1/\tau \ll 2\Delta$, while the dirty limit is $1/\tau > 2\Delta$. The clean and dirty limits may also be expressed as $l \gg \xi_0$ and $l < \xi_0$, respectively, where l is the quasiparticle mean-free path and ξ_0 is the BCS coherence length; because $l \propto \tau$ and $\xi_0 \propto 1/\Delta$, this is equivalent to the previous statement.[29] The use of these defintions depends on having accurate values for $1/\tau$ and Δ. In general, BCS superconductors have relatively low values for T_c, thus $1/\tau$ is assumed to have little temperature de-

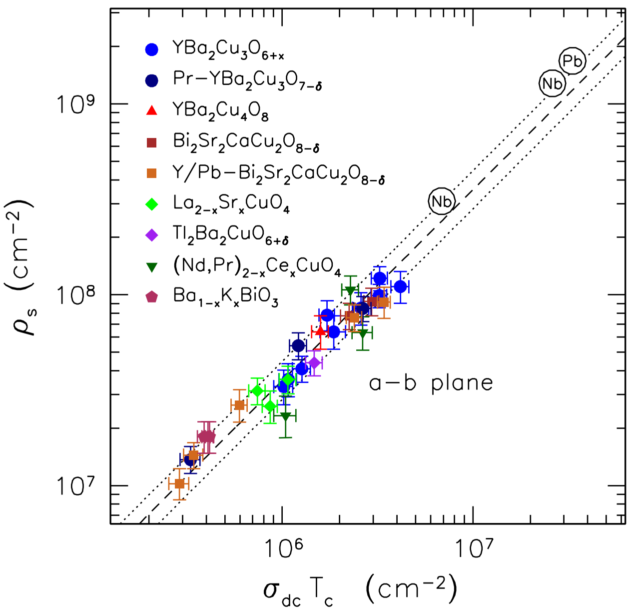

FIG. 1: The log-log plot of the superfluid density ρ_s vs $\sigma_{dc} T_c$ for the *a-b* planes of the hole-doped copper-oxide superconductors for pure and Pr-doped $YBa_2Cu_3O_{6+x}$ (Refs. 11,16,17,18,19); $YBa_2Cu_4O_8$ (Ref. 17); pure and Y/Pb-doped $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Refs. 19,20); underdoped $La_{2-x}Sr_xCuO_4$ (Ref. 21); $Tl_2Ba_2CuO_{6+\delta}$ (Ref. 22); electron-doped $(Nd,Pr)_{2-x}Ce_xCuO_4$ (Refs. 23,24,25) and the bismate material $Bi_{1-x}K_xBiO_3$ (Ref. 26). Within error, all the points may be described by a single (dashed) line. The points for Nb and Pb, indicated by the atomic symbols, also fall close to this line (Refs. 27,28).

pendence close to the superconducting transition. This assumption may be tested by suppressing T_c through the application of a magnetic field in excess of the upper critical field (H_{c2}) and examining the transport properties, which typically reveal little temperature dependence in $1/\tau$ below the zero-field value of T_c. The application of the the clean and dirty-limit picture to the copper-oxide superconductors is complicated by both the high critical temperature, and the superconducting energy gap which is thought to be *d*-wave in nature and momentum dependent (Δ_k), containing nodes.[30,31] The high value for T_c suggests that $1/\tau$ may still have a significant temperature dependence close to T_c. Indeed, below T_c the quasiparticle scattering rate in the cuprates is observed to decrease by nearly two orders of magnitude at low temperature.[32] This rapid decrease in $1/\tau$ is also observed optically, but not to the same extent.[33] A gap with $d_{x^2-y^2}$ symmetry may be written as $\Delta_k = \Delta_0 [\cos(k_x a) - \cos(k_y a)]$, where Δ_0 is the gap maximum. The fact that the scattering rate of the quasiparticles restricted to the nodal regions of the Fermi surface for $T \ll T_c$ is quite small has been taken as evidence that these materials are in the clean limit.[34,35,36] While it is certainly true that for $T \ll T_c$ the scattering rate is small and the nodal quasiparticles have very long mean-free paths, it is problematic to assert that the superconductor is therefore in the clean limit. In a normal BCS superconductor $1/\tau$ is also observed to decrease dramatically below T_c, regardless of the normal-state value of $1/\tau$, due to the formation of a condensate.[37]. Thus, the criteria of a small value of the quasiparticle scattering rate for $T \ll T_c$ is not a good measure of whether or not the superconductivity is in the clean or dirty limit. As with BCS materials, it is desirable to suppress T_c in the copper-oxide materials through the application of a magnetic field to determine the low-temperature behavior of $1/\tau$. While H_{c2} is quite large in the cuprates, experiments using pulsed magnetic fields can suppress T_c; in these experiments the resistivity of the optimally-doped materials matches the zero-field values at high temperatures due to the low magnetoresistance of these materials, and the trend of slowly decreasing resistivity continues smoothly to low temperatures,[38,39,40,41,42] often saturating at a value close to that observed at T_c. The implication of these experiments is that the normal-state value of $1/\tau$ is a good measure of the scattering rate in those systems in which T_c has been suppressed, and is therefore the value that should be considered when determining whether a system is in the clean or dirty limit. In addition to this explicit approach, a simpler method is to adopt an operational definition which states that if ρ_s changes with respect to the normal-state value of $1/\tau$ then the material is in the dirty limit; when ρ_s is no longer sensitive to the value of $1/\tau$ then the material is in the clean limit. Most of the materials in Fig. 1 are studied as a function of carrier doping, but it is also important to note that the introduction of disorder for fixed doping levels has also been studied.[16] The fact that all the observed results follow this linear scaling relation strongly suggests that the copper-oxide superconductors are close to or in the dirty limit (i.e., the superfluid density changes in response to variations in $1/\tau$).

Clean limit. The BCS model is used to describe the superconductivity of simple metals and alloys. If the normal-state properties may be described by the simple Drude model where the complex dielectric function is written as $\tilde{\epsilon}(\omega) = \epsilon_\infty - \omega_p^2/[\omega(\omega + i\gamma)]$, where $\omega_p^2 = 4\pi n e^2/m^*$ is the classical plasma frequency with the free-carrier concentration n and effective mass m^*, $\gamma = 1/\tau$ is the scattering rate, and ϵ_∞ is a high-frequency contribution. The dielectric function and the conductivity are related through $\tilde{\sigma} = \sigma_1 + i\sigma_2 = -i\omega\tilde{\epsilon}/4\pi$, thus the frequency-dependent conductivity has the form $\sigma_1(\omega) = \sigma_{dc}/(1 + \omega^2\tau^2)$ and $\sigma_{dc} = \omega_p^2\tau/4\pi$, which has the shape of a Lorentzian centered at zero frequency with a width at half-maximum given by $1/\tau$. The optical conductivity below T_c has been calculated from an isotropic (*s*-wave) energy gap 2Δ that considers an arbitrary purity level.[43] The clean limit case ($1/\tau \ll 2\Delta$) is illustrated in Fig. 2 for the choice $1/\tau = 0.2\Delta$. An aspect of clean-limit systems is that nearly all of the spectral weight associated with the condensate lies below 2Δ. As a result, the normalized spectral weight of the condensate[44] $(N_n - N_s)/\rho_s$ shown in the inset of Fig. 2

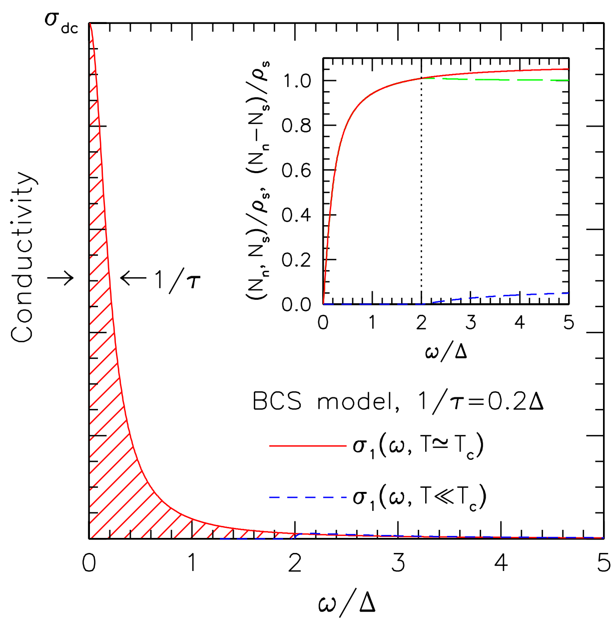

FIG. 2: The optical conductivity for the BCS model in the normal and superconducting states for a material in the clean limit ($1/\tau \ll 2\Delta$). The normal-state conductivity is a Lorentzian centered at zero frequency with a full width at half maximum of $1/\tau$ for $T \simeq T_c$ (solid line); the conductivity for $T \ll T_c$ is only faintly visible (dashed line). The spectral weight associated with the formation of a superconducting condensate is indicated by the hatched area. Inset: $N_n = N(\omega, T \simeq T_c)$ (solid line), $N_s = N(\omega, T \ll T_c)/\rho_s$ (dashed line), and difference between the two (long-dashed line), normalized with respect to ρ_s; $(N_n - N_s)/\rho_s$ converges rapidly to unity, and is fully formed at energies comparable to $1/\tau$.

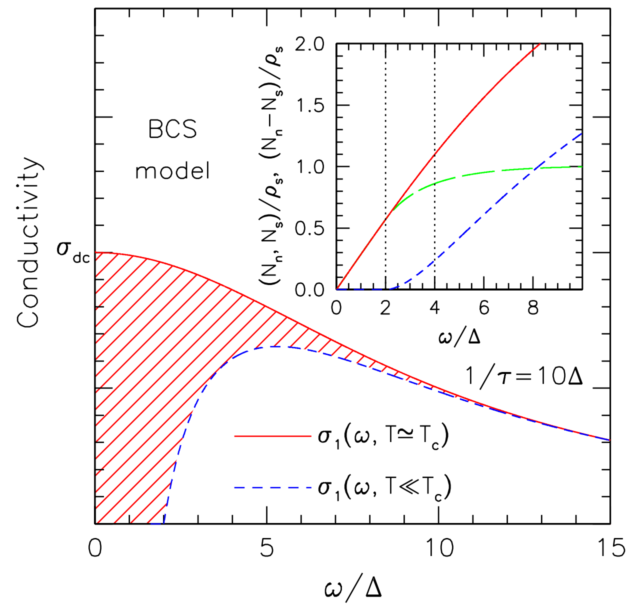

FIG. 3: The optical conductivity for the BCS model in the normal (solid line) and superconducting states (dashed line) for a material in the dirty limit ($1/\tau > 2\Delta$). The spectral weight associated with the formation of a superconducting condensate is indicated by the hatched area. Inset: $N_n = N(\omega, T \simeq T_c)$ (solid line), $N_s = N(\omega, T \ll T_c)$ (short dashed line), and difference between the two (long dashed line), normalized with respect to ρ_s; $(N_n - N_s)/\rho_s$ converges to unity at energies comprable to 2Δ.

approaches unity at frequencies closer to $1/\tau$ rather than 2Δ. The spectral weight for the condensate (the difference in the area under the two curves, indicated by the hatched region) may be estimated as $\rho_s \simeq \sigma_{dc}/\tau$. If $1/\tau \propto T_c$ for $T \simeq T_c$ in the copper-oxide materials,[45] then $\rho_s \propto \sigma_{dc} T_c$, in agreement with the observed scaling relation. It is interesting to note that $1/\tau \propto T_c$ yields rather large values for the normal-state scattering rate, and it has been suggested that the copper-oxide materials are close to the maximum level of dissipation allowed for these systems.[46] Furthermore, even though a d-wave system complicates the interpretation of the clean and dirty limit, large normal-state values of $1/\tau$ and relatively short normal-state mean-free paths[47] are problematic for a clean-limit picture; to achieve the clean limit it is not only necessary that $1/\tau \ll 2\Delta_0$, but also that $1/\tau \lesssim 2\Delta_k$ in the nodal regions. In fact, the clean-limit requirement is much more stringent for a d-wave system than it is for a material with an isotropic energy gap, and it is not clear that it will ever be satisfied in the copper-oxide superconductors. This suggests that a dirty-limit view may be more appropriate.

Dirty limit. In the BCS dirty limit, $1/\tau > 2\Delta$; this is illustrated in Fig. 3 for $1/\tau = 10\Delta$. In this case the normal-state conductivity is a broadened Lorentzian, and much of the spectral weight has been pushed out above 2Δ. As a result, the normalized spectral weight of the condensate, shown in the inset, converges much more slowly than in the clean-limit case. However, a majority of the spectral weight is captured by 2Δ and ρ_s is almost fully formed above 4Δ (Ref. 44). In the dirty-limit case, the spectral weight of the condensate (the hatched area in Fig. 3) may be estimated as $\rho_s \simeq \sigma_{dc}\, 2\Delta$. In the BCS model, the energy gap 2Δ scales linearly with T_c, yielding $\rho_s \propto \sigma_{dc} T_c$, which is in agreement with the observed scaling relation. As in the clean-limit case, the nature of the gap is important. However, if $1/\tau > 2\Delta_0$, the spirit of the dirty-limit case is preserved for all Δ_k. While many of the points in Fig. 1 are doping-dependent studies and do not track systematic changes in $1/\tau$, some of these points are for the same chemical doping with different scattering rates resulting from disorder that has either been deliberately introduced,[16] or that exist simply as a byproduct of synthesis.[48,49] The observation that all the points obey a linear scaling relation satisfies the operational definition of the dirty limit, suggesting that the examined materials are either close to or in the dirty limit.

It was noted in Fig. 1 that the points for Nb and Pb agreed reasonably well with the scaling relation used to

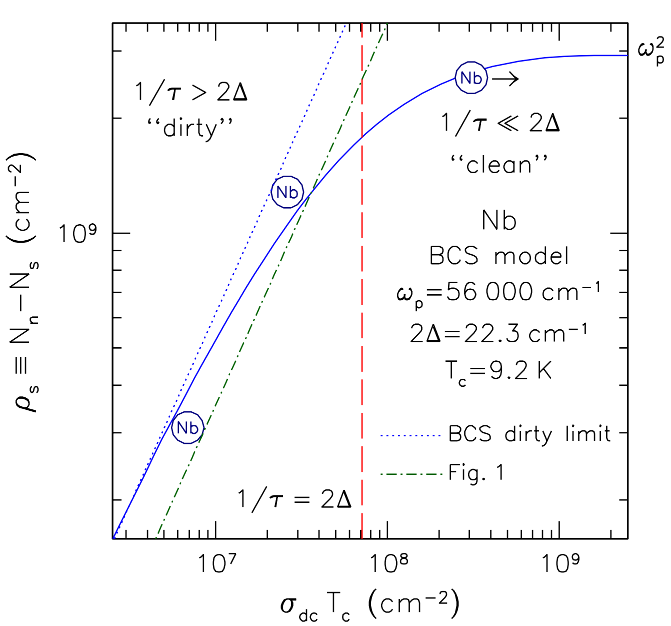

FIG. 4: The log-log plot of the predicted behavior from the BCS model of the strength of the condensate for Nb for a wide range of scattering rates $1/\tau = 0.05\Delta \rightarrow 50\Delta$, and assuming a plasma frequency $\omega_p = 56\,000$ cm^{-1}, critical temperature $T_c = 9.2$ K and an energy gap of $2\Delta = 3.5\,k_B\,T_c$ (solid line). The dashed line indicates $1/\tau = 2\Delta$. To the right of this line the material approaches the clean limit with a residual resistance ratio (RRR) of $\gtrsim 100$; the right arrow indicates that for larger RRR's, σ_{dc} close to T_c increases, but ρ_s has saturated to ω_p^2 (the data point for Nb in this regime is from Ref. 50). As the scattering rate increases, the strength of the condensate adopts a linear scaling behavior (dotted line); the two points for Nb (Refs. 27,28) shown in Fig. 1 lie close to this line, indicating that they are in the dirty limit. The scaling relation shown in Fig. 1 (dash-dot line) is slightly offset from the BCS dirty-limit result.

describe the copper-oxide superconductors. It is important to determine if these values represent clean or dirty-limit results. The expected behavior of Nb has been modeled using the BCS model[43] with a critical temperature of $T_c = 9.2$ K and a gap of $2\Delta = 22.3$ cm^{-1} (the BCS weak-coupling limit $2\Delta = 3.5\,k_B\,T_c$). The normal-state is described using the Drude model with a classical plasma frequency of $\omega_p = 56\,000$ cm^{-1} (Ref. 51) and a range of scattering rates $1/\tau = 0.05\Delta \rightarrow 50\Delta$; from the Drude model the dc conductivity is $\sigma_{dc} = \omega_p^2\,\tau/60$ (in units of Ω^{-1}cm^{-1} when the plasma frequency and the scattering rate have units of cm^{-1}). The spectral weight of the condensate $\rho_s = N_n - N_s$ has been determined by integrating to $\omega \simeq 200\Delta$, where ρ_s is observed to converge for all the values of $1/\tau$ examined. The result of this calculation is shown as the solid line in Fig. 4, and the dashed line indicates where $1/\tau = 2\Delta$. The point to the right of the dashed line is for Nb recrystallized in ultra-high vacuum[50] to achieve clean-limit conditions in which the residual resistivity ratios ($\rho_{\rm RT}/\rho_{T \gtrsim T_c}$) are in excess of 100, and where $\rho_s \rightarrow \omega_p^2$ for $T \ll T_c$. As the scattering rate increases and the material becomes progressively more "dirty", the strength of the condensate begins to decrease until it adopts the linear scaling behavior $\rho_s \simeq 60\,\sigma_{dc}\,T_c$ observed in Fig. 4. (It should be noted that the BCS model yields the same asymptotic behavior in the dirty limit, regardless of the choice of ω_p or Δ; the constant is only sensitive upon the ratio of Δ to T_c.) The two points for Nb shown in Fig. 1, (reproduced in Fig. 4), fall close to this line[27,28] and are clearly in the dirty limit. Thus, the scaling relation $\rho_s \propto \sigma_{dc}\,T_c$ is the hallmark of a BCS dirty-limit system. The scaling relation for the copper-oxide superconductors $\rho_s \simeq 35\,\sigma_{dc}\,T_c$ is slightly less than the asymptotic behavior observed for the weak-limit BCS material. This difference is perhaps due to the different symmetry of the superconducting energy gap in the two systems, and the fact that in the copper-oxide materials there is still a substantial amount of low-frequency residual conductivity at low temperature. Regardless of these differences, the empirical scaling relation $\rho_s \propto \sigma_{dc}\,T_c$ is observed in both the copper oxide and disordered elemental superconductors. If it is true in general that $\rho_s \propto \sigma_{dc}\,2\Delta$, then this necessarily implies that $\Delta \propto T_c$. In the optimally-doped and overdoped materials, there is some evidence that $\Delta_0 \propto T_c$ (Ref. 52). In the underdoped materials, large gaps are observed to develop in the normal state[53] well above T_c. While it has been observed that the energy scale over which spectral weight is transferred into the condensate is much larger in the underdoped materials than it is for the optimally-doped materials,[54,55] the majority of the spectral weight is still captured at energies comparable to T_c. This would support the view that the energy scale relevant to the formation of the condensate is proportional to T_c.

It is of some interest at this point to compare the empirical relation, that ρ_s is proportional to $\sigma_{dc}\,T_c$, with the expression for the penetration depth that is given by the Ginzburg-Landau theory modified for the dirty limit. In general, the expression for the London penetration depth is given by $\lambda_L(T \rightarrow 0) = \sqrt{mc^2/(4\pi n_s e^2)}$, where $n_s \equiv n$ is the superconducting carrier concentration. In the dirty limit one can show that $\rho_s(\text{dirty})/\rho_s(\text{clean}) = l/\xi_0$ (Ref. 29). An increase in $1/\tau$ reduces the amount of superfluid and the penetration depth increases and can be written as $\lambda^2 = (\xi_0/l)\,\lambda_L^2$. Since $\lambda^2 \propto 1/\rho_s$, $\xi_0 \propto 1/T_c$, and $\sigma_{dc} \propto l$, then one can recover the result that $\rho_s \propto \sigma_{dc}\,T_c$. It is possible that in a *d*-wave system the presence of nodal regions with a small superfluid density and $\Delta_k \ll \Delta_0$, that the coherence length in the above expression for λ^2 now involves some average including the nodal regions.

Summary. The implications of the linear scaling relation $\rho_s \propto \sigma_{dc}\,T_c$ in the copper-oxide superconductors has been examined within the context of clean and dirty-limit systems. In the conventional BCS superconductors (such as Nb), this linear scaling is the hallmark of a dirty-limit system. The copper-oxide materials are thought to be *d*-

wave superconductors, in which the clean limit is difficult to achieve. The observed linear scaling strongly suggests that the copper-oxide superconductors are either close to or in the dirty limit. Estimates of ρ_s based on geometric arguments imply that the energy scale below which the majority of the spectral weight is transferred into the condensate scales linearly with T_c.

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* Electronic address: homes@bnl.gov

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