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# Scaling of the superfluid density in high-temperature superconductors

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#### Abstract

A scaling relation  $N_c \simeq 4.4\sigma_{dc}T_c$  has been observed parallel and perpendicular to the copper-oxygen planes in the high-temperature superconductors;  $N_c$  is the spectral weight and  $\sigma_{dc}$  is the dc conductivity just above the critical temperature  $T_c$ . In addition, Nb and Pb also fall close to this scaling line. The application of the Ferrell-Glover-Tinkham sum rule to the BCS optical properties of Nb above and below  $T_c$  yields  $N_c \simeq 8.1\sigma_{dc}T_c$  when the normal-state scattering rate is much greater than the superconducting energy gap  $(1/\tau > 2\Delta)$ , the "dirty" limit). This result suggests that the high-temperature superconductors may be in the dirty limit. The superconductivity perpendicular to the planes is explained by the Josephson effect, which again yields  $N_c \simeq 8.1\sigma_{dc}T_c$  in the BCS formalism. The similar forms for the scaling relation in these two directions suggests that in some regime the dirty limit and the Josephson effect may be viewed as equivalent.

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## 1. Introduction

Scaling laws express a systematic and universal simplicity in nature. Since the discovery of superconductivity at elevated temperatures in copper-oxide materials some 20 years ago [1], there has been considerable effort to identify trends and correlations between the physical quantities as a clue to the origin of the superconductivity [2]. One of the earliest patterns that emerged was the linear scaling of the superfluid density  $\rho_s$  in the copper–oxygen planes of the hole-doped materials with the superconducting transition temperature  $T_c$  (where  $\rho_s$  is proportional to the number of carriers in the condensate  $n_s$ ; in addition  $\rho_s \propto 1/\lambda^2$ , where  $\lambda$  is the superconducting penetration depth). This is the celebrated Uemura relation [3,4] and it works well for the underdoped materials. However, this relation appears to break down in the very underdoped [5] and

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overdoped [6,7] materials (optimal doping is defined where  $T_c$  is a maximum [8]).

In contrast, we have recently demonstrated a scaling relation  $N_c \simeq 4.4 \sigma_{dc} T_c$  [9–11], where  $N_c$  is the spectral weight of the condensate ( $N_c = \rho_s/8$ ) and  $\sigma_{dc}$  is the dc conductivity just above the critical temperature. (In this instance both sides of the equation possess the same units, so that the constant is dimensionless. The dimensionless constant and the description of the scaling in terms of  $N_{\rm c}$ rather than  $\rho_s$  results in a prefactor which is smaller than observed in our previous work [9]). This relation appears to hold regardless of the doping level or type, nature of the disorder, or direction (along or perpendicular to the copper-oxygen planes). In addition to the copper-oxide materials, the simple elemental BCS superconductors Nb and Pb are also observed to follow this relation. The optical properties of Nb were calculated in the normal and superconducting states. The spectral weight  $N_c$  is then determined from conductivity sum rules. The linear scaling  $N_{\rm c} \propto \sigma_{\rm dc} T_{\rm c}$  is recovered in the BCS "dirty" limit  $1/\tau > 2\Delta$ , where  $1/\tau$  is the normal-state scattering rate and  $2\Delta$  is the

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isotropic BCS superconducting energy gap ( $T \ll T_c$ ). This result suggests that the copper-oxide materials may be in the "dirty" limit. The superconductivity perpendicular to the planes is thought to arise from Josephson coupling; interestingly, this approach again yields the scaling relation  $N_c \propto \sigma_{dc} T_c$  [10]. This result and a possible connection with the in-plane behavior is discussed.

#### 2. Experiment and results

The relevant experimental quantities here are the spectral weight of the condensate  $(N_c \propto 1/\lambda^2)$  for  $T \ll T_c$  and  $\sigma_{\rm dc}$  at  $T \simeq T_{\rm c}$ . Normally, these two quantities are determined using different experimental techniques on different samples where the dopings are at best similar, but never identical. A fundamental advantage of optical reflectance techniques is that the real and imaginary parts of the dielectric function  $\tilde{\epsilon} = \epsilon_1 + i\epsilon_2$  may be determined, allowing both  $N_{\rm c}$  and  $\sigma_{\rm dc}$  to be calculated for the same sample. The reflectance of a large number of single and double layer cuprates has been measured over a wide frequency range by a number of different workers, and the Kramers-Kronig analysis used to calculate the complex optical properties [11]. The real part of the optical conductivity is  $\sigma_1(\omega) = -i\omega\epsilon_2/4\pi$ (in units of cm<sup>-1</sup>), and  $\sigma_{dc} = \sigma_1(\omega \to 0)$  at  $T \simeq T_c$ . For  $T \ll T_{\rm c}$ , the response of  $\tilde{\epsilon}$  to the formation of a superconducting condensate is ideally purely real, thus  $\epsilon_1 =$  $\epsilon_{\infty} - \omega_{\rm ps}^2/\omega^2$ , and  $\omega_{\rm ps}^2 = -\omega^2 \epsilon_1(\omega)$  in the  $\omega \to 0$  limit. Here,  $\omega_{\rm ps}^2 = 4\pi n_s e^2/m^*$  is the square of the superconducting plasma frequency,  $n_{\rm s}$  is the superconducting carrier concentration,  $m^*$  is an effective mass, and  $\epsilon_{\infty}$  is the high-frequency contribution to the real part of the dielectric function. The strength of the condensate is simply  $\rho_{\rm s} \equiv \omega_{\rm ps}^2$ , which is proportional to  $n_{\rm s}/m^*$ . The value of  $\rho_{\rm s}$ may also be estimated by examining the changes in the optical conductivity just above and well below  $T_{\rm c}$ . The *f*-sum rule for the conductivity [12] has the form  $\int_0^\infty \sigma_1(\omega) d\omega = \omega_p^2/8$ , where  $\omega_p^2 = 4\pi n e^2/m$  is the classical plasma frequency. The spectral weight at a given cut-off frequency  $\omega_c$  is defined here as

$$N(\omega_{\rm c},T) = \int_{0^+}^{\omega_{\rm c}} \sigma_1(\omega,T) \mathrm{d}\omega, \qquad (1)$$

which is simply the area under the conductivity curve. The copper-oxide materials, and superconductors in general, show a dramatic suppression of the low-frequency conductivity upon entering the superconducting state; this difference between the  $T \simeq T_c$  and  $T \ll T_c$  conductivities is often referred to as the "missing area". The spectral weight associated with the formation of the superconducting condensate is then  $N_c = N_n - N_s$ , where  $N_n \equiv N(\omega_c, T \simeq T_c)$ , and  $N_s \equiv N(\omega_c, T \ll T_c)$ , and  $\omega_c$  is chosen such that  $N_c$  converges. Here,  $N_c$  is simply the spectral weight associated with the missing area in the conductivity, which is related to the square of the superconducting plasma frequency

$$\omega_{\rm ps}^2 = 8N_{\rm c},\tag{2}$$

or  $\rho_s = 8N_c$ . This expression is the well-known Ferrell– Glover–Tinkham sum rule [13,14]. These two different techniques typically yield nearly identical values for  $\rho_s$ ; an exception exists in the underdoped materials perpendicular to the planes, where it has been suggested that there is missing spectral weight [15]. The optically-determined values of  $\omega_{ps}$  and  $\sigma_{dc}$  [16] for a wide variety of copper–oxygen superconductors are listed in Table I of Ref. [11].

# 3. Discussion

In order to determine whether or not the Uemura relation is appropriate for the optical data, a plot of  $N_c$  vs  $T_c$  for the copper-oxide superconductors was investigated, shown in the log-log plot in Fig. 1. While the data points that characterize the underdoped materials follow the  $N_c \propto T_c$  relation (the dashed line in Fig. 1), many of the optimally and overdoped materials do not. The electron-doped materials in particular present a serious problem as they fall well off the scaling line. In contrast, a plot of  $N_c$  vs  $\sigma_{dc}T_c$  shown in Fig. 2 indicates that, within error, all of the points may be described by the relation  $N_c \simeq 4.4\sigma_{dc}T_c$ .

A surprising result in Fig. 2 is that in addition to the copper-oxide superconductors, materials such as Pb and Nb also fall very close to the scaling line. The copper-oxide materials are thought to possess gaps which are d wave in nature and contain nodes [17,18]. A gap with  $d_{r^2-v^2}$ 



Fig. 1. The log-log plot of the spectral weight of the condensate  $N_c$  vs  $T_c$  of the hole-doped copper-oxide superconductors for pure and Pr-doped YBa<sub>2</sub>Cu<sub>3</sub>O<sub>6+x</sub>; pure and Zn-doped YBa<sub>2</sub>Cu<sub>4</sub>O<sub>8</sub>; pure and Y/Pb-doped Bi<sub>2</sub>Sr<sub>2</sub>CaCu<sub>2</sub>O<sub>8+ $\delta$ </sub>; underdoped La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub>; Tl<sub>2</sub>Ba<sub>2</sub>CuO<sub>6+ $\delta$ </sub>; electron-doped (Nd,Pr)<sub>2-x</sub>Ce<sub>x</sub>CuO<sub>4</sub> and the bismuth oxide material Bi<sub>1-x</sub>K<sub>x</sub>BiO<sub>3</sub>. The underdoped materials follow the  $N_c \propto T_c$  relation reasonably well (dashed line); however, the optimally and overdoped materials, as well as the electron-doped systems, deviate substantially from this line. (The values used in the plot are shown in Table I of Ref. [11].)



Fig. 2. The log–log plot of the spectral weight of the condensate  $N_c$  vs  $\sigma_{dc}T_c$  for the same materials shown in Fig. 1. Within error, all the points may be described by a single (dashed) line,  $N_c \simeq 4.4\sigma_{dc}T_c$ ; the upper and lower dotted lines represent approximately the spread of the data.

symmetry may be written as  $\Delta_k = \Delta_0 [\cos(k_x a) - \cos(k_y a)];$ the gap reaches a maximum at the  $(0,\pi)$  and  $(\pi,0)$  points, and vanishes along the nodal  $(\pi,\pi)$  directions. On the other hand, metals such as Pb and Nb are BCS superconductors which have nearly isotropic s-wave energy gaps. The two systems might reasonably be expected to follow different scaling relations. A deeper understanding of the scaling relation as it relates to BCS superconductors, and possibly the copper-oxide materials, may be obtained from an examination of the spectral weight above and below  $T_{\rm c}$ as determined by the normal-state scattering rate and an isotropic superconducting energy gap. In order to achieve this, the optical properties for Nb have been calculated in both the normal and superconducting states. The "metallic" normal state may be described by the Drude model where the complex dielectric function is

$$\tilde{\epsilon}(\omega) = \epsilon_{\infty} - \frac{\omega_{\rm p}^2}{\omega(\omega + \mathrm{i}\gamma)},$$
(3)

 $\epsilon_{\infty}$  and the  $\omega_{\rm p}$  have been previously defined, and  $\gamma = 1/\tau$ . The dielectric function and the conductivity are related through  $\tilde{\sigma} = \sigma_1 + i\sigma_2 = -i\omega(\tilde{\epsilon} - \epsilon_{\infty})/4\pi$ . Thus  $\sigma_1 = \sigma_{\rm dc}/(1 + \omega^2\tau^2)$  with  $\sigma_{\rm dc} = \omega_{\rm p}^2\tau/4\pi$  (in units of cm<sup>-1</sup>), which has the shape of a Lorentzian centered at zero frequency, with a width of  $1/\tau$ . The plasma frequency for Nb has been taken to be  $\omega_{\rm p} = 56000 \,{\rm cm}^{-1}$  [19]. The behavior of Nb in the superconducting state has been calculated using the BCS model [20] for an arbitrary purity level with a critical temperature of  $T_{\rm c} = 9.2 \,{\rm K}$  and a gap of  $2\Delta = 22.3 \,{\rm cm}^{-1}$ (the BCS weak-coupling limit  $2\Delta = 3.5k_{\rm B}T_{\rm c}$ ); a wide range of normal-state scattering rates  $1/\tau = 0.05\Delta \rightarrow 50\Delta$  have been examined. The spectral weight of the condensate  $N_{\rm c}$ has been determined by integrating to  $\omega_c \simeq 200 \Delta$ ; N<sub>c</sub> is observed to converge smoothly for all the values of  $1/\tau$  examined. The result of this calculation is shown as the solid line in Fig. 3, and the vertical 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 [21] to achieve conditions in which the residual resistivity ratios  $\left[\rho(RT)\right]$  $\rho(T \gtrsim T_{\rm c})$ ] are well in excess of 100, and where  $N_{\rm c} \rightarrow$  $\omega_{\rm p}^2/8$  (or  $\rho_{\rm s} \to \omega_{\rm p}^2$ ) for  $T \ll T_{\rm c}$ . As the scattering rate increases the strength of the condensate begins to decrease until it adopts the linear scaling behavior  $N_{\rm c} \simeq 8.1 \sigma_{\rm dc} T_{\rm c}$ observed in Fig. 3. (It should be noted that the BCS model yields the same asymptotic behavior in the dirty limit, regardless of the choice of  $\omega_p$  or  $\Delta$ ; the constant is only sensitive upon the ratio of  $\Delta$  to  $T_{\rm c}$ .) The two points for Nb shown in Fig. 2 [22,23], (reproduced in Fig. 3), fall close to this line and are clearly in the dirty limit. Thus, the scaling relation  $N_{\rm c}$  or  $\rho_{\rm s} \propto \sigma_{\rm dc} T_{\rm c}$  is the hallmark of a BCS dirty-limit system [24]. The presence of  $\sigma_{dc}$  in the scaling relation indicates the nature of the superconductivity depends on the normal-state scattering rate. To illustrate this



Fig. 3. The log-log plot of the predicted behavior from the BCS model of the spectral weight of the condensate  $N_c$  in Nb for a wide range of scattering rates  $1/\tau = 0.05 \Delta \rightarrow 50 \Delta$ , and assuming a plasma frequency  $\omega_p = 56000 \text{ cm}^{-1}$ , critical temperature  $T_c = 9.2$  K and an energy gap of  $2\Delta = 3.5k_BT_c$  (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,  $\sigma_{dc}$  close to  $T_c$  increases, but  $N_c$  has saturated to  $\omega_p^2/8$  (or  $\rho_s \rightarrow \omega_p^2$ ; the data point for Nb in this regime is from Ref. [21]). As the scattering rate increases, the spectral weight of the condensate adopts a linear scaling behavior (dotted line); the two points for Nb (Refs. [22,23]) shown in Fig. 2 lie close to this line, indicating that they are in the dirty limit. The scaling relation shown in Fig. 2 (dash-dot line) is slightly offset from the BCS dirty-limit result.

point more clearly, we consider the two extreme limits in Fig. 3; the clean and dirty limits.

The clean limit case  $(1/\tau \ll 2\Delta)$  is illustrated in Fig. 4 for the choice  $1/\tau = 0.2\Delta$ . Nearly all of the spectral weight associated with the condensate lies below  $2\Delta$ . As a result, the normalized spectral weight of the condensate  $8N_c/\rho_s$ (the difference in the area under the two curves indicated by the hatched region) shown in the inset of Fig. 4, approaches unity at frequencies closer to  $1/\tau$  rather than  $2\Delta$ . The spectral weight for the condensate may be estimated simply as a geometric area  $N_{\rm c} \simeq \sigma_{\rm dc}/\tau$ . If  $1/\tau \propto T_{\rm c}$ for  $T \simeq T_c$  in the copper-oxide materials [25], then  $N_{\rm c} \propto \sigma_{\rm dc} T_{\rm c}$ , in agreement with the observed scaling relation. It is interesting to note that  $1/\tau \propto T_c$  may yield 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 [26]. In addition, to achieve the clean limit it is not only necessary that  $1/\tau \ll 2\Delta_0$ , but also that  $1/\tau \leq 2\Delta_k$  in the nodal regions. 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. It should be emphasized at this point that in the high-temperature superconductors the large normal-state scattering rate is not due to impurities, but rather from out-of-plane disorder, correlation effects, or possibly a combination of both.

The BCS dirty limit  $(1/\tau > 2\Delta)$  is shown in Fig. 5 for the choice of  $1/\tau = 10\Delta$ . In this case the normal-state conductivity is a broadened Lorentzian, and much of the spectral weight has been pushed out above  $2\Delta$ . 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  $2\Delta$  and  $N_c$  is almost fully formed above  $4\Delta$ . In this case, the spectral weight of the condensate (the hatched area in Fig. 5) may be estimated as  $N_c \simeq \sigma_{dc} 2\Delta$ . In the BCS model, the energy gap  $2\Delta$  scales linearly with  $T_c$ , yielding  $N_c \propto \sigma_{dc} T_c$ , which is in agreement with the observed scaling relation. This result necessarily implies that the energy scale for the condensate is proportional to  $T_c$ .

The scaling relation predicted by the BCS model has a numerical constant of 8.1, but the geometrical estimate assuming weak coupling yields a value of only 2.4. An examination of Fig. 5 indicates that this discrepancy arises from the fact that  $N_c \simeq \sigma_{dc} 2\Delta$  underestimates the spectral weight by more than a factor of two. The results from Fig. 3 suggest that a more realistic estimate of the area is  $N_c \simeq 3.3(\sigma_{dc} 2\Delta)$ , which assuming weak coupling yields the correct numerical constant in the scaling relation. The observed scaling relation in the cuprates would imply that  $2\Delta/k_BT_c \simeq 2$ . However, this statement suffers from the fact

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BCS model





Conductivity 0.5 ß d 0 2 6 8 4  $\omega / \Delta$ ΟΔ  $(\omega, \top \simeq \top)$  $_{1}(\omega, \top \ll \top_{c})$ 15  $\omega / \Delta$ Fig. 5. The optical conductivity for the BCS model in the normal (solid

Fig. 5. The optical conductivity for the BCS model in the normal (solid line) and superconducting states (dashed line) for a material in the dirty limit ( $1/\tau \gtrsim 2\Delta$ ). The spectral weight associated with the formation of a superconducting condensate is indicated by the hatched area. A significant amount of spectral weight lies above  $2\Delta$ . Inset:  $N_n \equiv N(\omega, T \simeq T_c)$  (solid line),  $N_s \equiv N(\omega, T \ll T_c)$  (dashed line), and difference between the two  $N_c = N_n - N_s$  (long-dashed line) normalized with respect to  $\rho_s/8$ ;  $8N_c/\rho_s$  converges at energies comparable to  $4\Delta$ .

that it is valid only within a BCS formalism for an isotropic s-wave gap. Another, perhaps more reasonable, explanation for the different numerical constants between the BCS and high-temperature superconductors may arise from the fact that copper-oxide superconductors have nodes in the energy gap, and as a consequence there is still a substantial amount of low-frequency residual conductivity at low temperature [27] resulting in a reduced estimate for the spectral weight for the condensate.

It was previously noted [9] that the scaling relation  $N_{\rm c} \simeq$  $4.4\sigma_{\rm dc}T_{\rm c}$  is a universal result that describes not only the copper-oxygen (a-b) planes, but perpendicular to the planes (*c*-axis) as well, as shown in Fig. 6. While a description of the scaling based on scattering rates within the context of clean and dirty limits may be appropriate for the a-b planes where the transport is coherent, it is inappropriate along the *c*-axis, where the activated nature of the temperature dependence of the resistivity indicates that the transport in this direction is incoherent and governed by hopping [28]. In this case, the superconductivity along the c-axis may be described by the Josephson effect, which for the BCS weak coupling case  $(2\Delta = 3.5k_{\rm B}T_{\rm c})$  yields  $N_{\rm c} \simeq 8.1 \sigma_{\rm dc} T_{\rm c}$  [10]. Surprisingly, this is precisely the result that was obtained in the *a*-*b* planes for the BCS weak-coupling case in the dirty limit in Fig. 3, indicating that from a functional point of view the scaling behavior of the dirty limit and the Josephson effect are nearly identical. Because these calculations have all been performed using a BCS for-



Fig. 6. The log–log plot of the spectral weight of the condensate  $N_c$  vs  $\sigma_{dc}T_c$  for the *a*-*b* planes and the *c*-axis for a variety of cuprates. Within error, all of the points fall on the same universal (dashed) line defined by  $N_c \simeq 4.4\sigma_{dc}T_c$ ; the dotted line is the dirty limit result  $N_c \simeq 8.1\sigma_{dc}T_c$  for the BCS weak-coupling case  $(2\Delta = 3.5k_BT_c)$  from Fig. 3, and also represents the Josephson result for the BCS weak-coupling case, used to describe the scaling along the *c*-axis [10]. The subscripts for the *c*-axis points are listed in the supplemental information of Ref. [9].).

malism, there is some uncertainty in applying these results to d-wave systems. It is possible that the Josephson effect arises naturally out of systems with an increasing amount of disorder and as a result any crossover from coherent to incoherent behavior still yields the same form of the scaling relation. The dynamical nature of the electronic inhomogeneities in the copper–oxygen planes may support this argument [29].

### 4. Conclusions

The implications of the linear scaling relation  $N_c$  or  $\rho_{\rm s} \propto \sigma_{\rm dc} T_{\rm c}$  in the copper-oxide superconductors have 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 superconductor. The copper-oxide materials are thought to be d-wave superconductors, in which the clean limit may be difficult to achieve. The observed linear scaling suggests that the copper-oxide superconductors may be close to or in the dirty limit. Estimates of  $N_{\rm c}$  (or  $\rho_{\rm s}$ ) 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  $T_c$ . The *a*-*b* planes and the *c*-axis are observed to follow the same scaling relation. The scaling behavior for the dirty limit and the Josephson effect (assuming a BCS formalism) is essentially identical from a functional point of view, suggesting that in some regime these two effects may be viewed as equivalent.

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