Scaling relation for the superfluid density in cuprates - origins and limits

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A universal scaling relation, \(\rho_s \propto \sigma(T_c) \times T_c\) has been reported by Homes \textit{et al.} (Nature (London) \textbf{430}, 539 (2004)) where \(\rho_s\) is the superfluid density and \(\sigma(T)\) is the DC conductivity. The relation was shown to apply to both \(c\)-axis and in-plane dynamics for high-\(T_c\) superconductors as well as to the more conventional superconductors Nb and Pb, suggesting common physics in these systems. We show quantitatively that the scaling behavior has several possible origins including, marginal Fermi-liquid behavior, Josephson coupling, dirty-limit superconductivity and unitary impurity scattering for a \(d\)-wave order parameter. However, the relation breaks down seriously in overdoped cuprates, and possibly even at lower doping.

The absence of a recognized theory of high-\(T_c\) superconductors (HTS) has led to a search for universal relationships that might provide a guide to the essential physics for HTS. The Uemura relation\(^{1,2}\) is one such scaling relation, namely \(\rho_s(0) \propto T_c\), where \(T_c\) is the superconducting (SC) transition temperature and \(\rho_s(0) = \lambda_{ab}^{-2} = \mu_0 e^2 n_s / m^*\) is the superfluid density. Here \(n_s\) is the density of SC electrons, \(m^*\) is its effective mass and \(\lambda_{ab}\) is the in-plane London penetration depth. The Uemura relation has been invoked in support of Bose-Einstein condensation of real-space pairs\(^3\),\(^4\) and is generally discussed as a test of theoretical models\(^4\). However, there is increasing recognition that this relation is an oversimplification\(^3\),\(^5\), and it breaks down on the overdoped side of the SC phase curve\(^5\).

Recently, a new scaling relation, \(\rho_s \propto \sigma(T_c) \times T_c\) was reported by Homes \textit{et al.}\(^8\) where \(\sigma(T)\) is the DC conductivity. This was shown to apply over five orders of magnitude including both \(c\)-axis and in-plane dynamics for HTS as well as to the conventional superconductors Nb and Pb. The authors suggested this relation may provide new insights into the origins of SC in HTS materials. We examine this relation and show that it is a natural consequence of several quite different but well-understood mechanisms, including dirty-limit conventional SC (Pb and Nb), Josephson coupling along the \(c\)-axis, marginal Fermi-liquid behavior (optimal and underdoped HTS), in-plane Josephson-coupled granular SC (strongly underdoped HTS) and Abrikosov-Gor’kov \(d\)-wave pair-breaking for non-magnetic scatterers. Since we submitted this work Homes \textit{et al.}\(^8\) have reported some overlapping results but here we examine the doping and impurity dependence of this relation showing that, in overdoped HTS, it breaks down seriously. It is perhaps significant that, here, the cuprates progress towards more conventional SC and normal state (NS) behavior.

Because \(\lambda^{-2} = \mu_0 e^2 n_s / m^*\) and \(\sigma = ne^2 \tau / m^*\) the correlation is equivalent to a relationship between scattering rate \(1/\tau\) and \(T_c\) given by \(h/\tau = 2.7 \pm 0.5 \times k_B T_c\). Here we assume that all the spectral weight associated with the free carriers condenses into the \(\delta\)-function at \(\omega=0\), i.e. \(n_s=n\). In fact, for underdoped cuprates some spectral weight remains at finite frequency\(^9\). However, we assume that this weight is related to some other excitation, different from the free carriers, which does not contribute to \(\sigma(\omega=0)\), \(\rho_s\) or the low-\(T\) specific heat.

In considering possible origins for this relation between \(h/\tau\) and \(k_B T_c\) we note that it is a direct prediction from Marginal-Fermi-liquid theory, where\(^{11,12}\)

\[
h/\tau \approx \pi k_B T_c + h\omega, \tag{1}\]

At low frequency, the scaling relation is almost exactly recovered provided \(\tau\) is evaluated at \(T_c\), as required.

That Nb and Pb fit the scaling line is surprising, but it is readily shown that for classical SC in the dirty limit \(\rho_s\) is, again, proportional to \(\sigma(T_c) \times T_c\). For a mean-free path, \(\ell\), and a BCS coherence length, \(\xi_0 = h v_F / (\pi \Delta)\), the effective penetration depth is given by\(^{13}\)

\[
\lambda_{eff} = \lambda_L (1 + \xi_0 / \ell)^{1/2} \approx \lambda_L (\xi_0 / \ell)^{1/2}. \tag{2}\]

Taking \(\ell = v_F \tau\) and \(\Delta = 1.76 k_B T_c\), we find that \(\rho_s\) is a factor of two higher than the scaling line \((h/\tau = 5.4 \times k_B T_c)\). In the clean limit there will be large deviations below the scaling line because \(\rho_s\) remains constant while \(\sigma(T_c) \times T_c\) can be extremely large. Using data for Pb alloys\(^{14}\) and for Nb alloys\(^{15}\) we find that, for the Pb sample on the Homes scaling line, \(\xi_0 / \ell = 1.2\) while the two Nb samples have \(\xi_0 / \ell = 0.6\) and 2.1. From eq. (2) it can be seen that in the intermediate situation where \(\xi_0 / \ell \approx 1\) the above factor of two is compensated. The apparent scaling here is understandable but fortuitous.

Homes \textit{et al.}\(^8\) consider Josephson coupling along the \(c\)-axis and show the proportionality \(\rho_s \propto \sigma \times T_c\) but do not...
evaluate the proportionality constant. The c-axis penetration depth, $\lambda_c$ is expressed in terms of the Josephson current density, $J_c$ as $\lambda_c^2 = h/(2\mu_0 d J_c)$ where $d$ is the spacing between SC planes and, for low temperatures, $J_c = \pi \Delta/(2e R_n)$. $R_n = d/\sigma_c$ is the NS tunnelling resistance between layers. For a $d$-wave gap we may assume that the effective gap parameter, $\Delta_{eff} = \Delta_0/\sqrt{2}$ where $\Delta_0 \approx 2.4 k_B T_c$ is the maximum $d$-wave gap near ($\pi, 0$). Thus $\lambda_c^2 = 6.79(\pi^2/4\alpha) k_B T_c \sigma_c$, giving the relation $h/\tau = 2.23 \times k_B T_c$. If instead we consider a simple rectangular I-V Josephson characteristic, then $I_c = 2\Delta_0/e R_n$ and the relation becomes $h/\tau = 2.83 \times k_B T_c$, again in excellent agreement with the scaling curve.

Thus, whether we consider Josephson coupling, dirty-limit SC or marginal-Fermi-liquid SC we recover the observed scaling behavior. Now we turn to the experimental data for the doping dependence of the in-plane dynamics.

In Fig. 1 the filled squares and diamonds show values of $\rho_s(0)$ plotted versus $\sigma(T_c) \times T_c$ for four different HTS materials. The samples, technique for measuring $\rho_s(0)$ and sample-type are as follows: (a) $Y_{1-x}$Ca$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ (transverse-field muon spin relaxation - $\mu$SR, polycrystalline), (b) Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ (field-dependent specific heat, polycrystalline), and (c) Tl$_2$Ba$_2$CuO$_{6+\delta}$ ($\mu$SR, polycrystalline) with La$_{2-x}$Sr$_x$CuO$_4$ (ac-susceptibility, oriented powders). The $\rho_s(0)$ data for Tl$_2$Ba$_2$CuO$_{6+\delta}$ are from ref. 6, and the remainder are from references in 8. The techniques noted are well established except for the specific-heat method. But $\rho_s(0)$ has also recently been measured for Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ using ac-susceptibility 17. Apart from the heavily underdoped region, the data are in excellent agreement with those used here from specific-heat.

The in-plane $\sigma(T)$ data for Fig. 1 are from DC transport measurements and for this it is essential to use single-crystal measurements. These are available for all cited samples except $Y_{1-x}$Ca$_x$Ba$_2$Cu$_3$O$_{7-\delta}$ for which we have used thin-film data from Fisher et al. supported by more recent data from Naqib et al. 19. Sources for the remaining samples are: Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ from Watanabe et al. 20; for Tl$_2$Ba$_2$CuO$_{6+\delta}$ from Tyler and coworkers 21 and for La$_{2-x}$Sr$_x$CuO$_4$ from Ando and coworkers 22. In all cases we evaluated $\sigma(T_c)$ at the reported doping states, fitted to an appropriate function of doping and interpolated to determine $\sigma(T_c)$ at the doping state relevant to the available $\rho_s(0)$ data. For Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$, the $\sigma(T_c)$ data had to be extrapolated to higher doping levels. In Fig. 1 optimal doping (maximal $T_c$) is indicated by the arrows and critical doping where the pseudogap closes, is the point of maximum $\rho_s$. The overdoped region lies further to the right.

Our main conclusion is that each compound exhibits a large departure from the “universal” scaling line in the overdoped region. In the optimal and underdoped regions, where we can directly compare with Homes et al., the situation is less clear. In some cases our data differs markedly from theirs which severely overestimates $\rho_s$ for Bi$_2$Sr$_2$CaCu$_2$O$_8$ relative to data from specific heat, ac susceptibility and $\mu$SR and underestimates $\rho_s$ for Tl$_2$Ba$_2$CuO$_6$. For this compound, and for La$_{2-x}$Sr$_x$CuO$_4$, their $\sigma(T_c)$ value is about half that observed from DC transport in single crystals 21. The source of these discrepancies may possibly be attributable to the extrapolation to $\omega = 0$ and $\omega = \infty$ required for the Kramers-Kronig transformation of the optical data.

Bearing in mind these differences our underdoped results show a consistent trend. For Bi$_2$Sr$_2$CaCu$_2$O$_8$, Tl$_2$Ba$_2$CuO$_6$ and La$_{2-x}$Sr$_x$CuO$_4$ the scaling line is not reached until near, or below, $1/8$th doping. On the other hand $Y_{1-x}$Ca$_x$Ba$_2$Cu$_3$O$_{7-\delta}$, though not linear, roughly follows the trend of the scaling line across the entire underdoped region. However, there are two reasons why this could misrepresent the situation. Firstly, compared with other bilayer cuprates with similar $T_c$ this compound generally exhibits a higher superfluid density arising from

![FIG. 1: Superfluid density, $\rho_s$ versus $\sigma(T_c) \times T_c$ for (a) $Y_{1-x}$Ca$_x$Ba$_2$Cu$_3$O$_{7-\delta}$, (b) Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ and (c) Tl$_2$Ba$_2$CuO$_{6+\delta}$ (overdoped only) and La$_{2-x}$Sr$_x$CuO$_4$. Arrows indicate optimal doping. In (a) the effects of Zn-induced pairbreaking are shown (crosses) along with the calculated A-G pairbreaking effects for constant $\sigma$ (dashed curve) and for $\sigma$ reduced by impurity scattering (dash-dot curve). The Homes’ data lie within $\pm 25\%$ of the scaling line.]

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the contribution of the chain layer. This may be approx-
imately balanced in the scaling plot by the additional
conductivity also arising from the chains and so we put
this effect to one side. But secondly, the conductivity
here is for thin films and thus is probably diminished rel-

tive to single-crystal data. All the data points may lie
further to the right, perhaps by a factor of the order of
1.5. Then the behavior would be rather similar to that
shown by Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$ and La$_{2-x}$Sr$_x$CuO$_4$. It may be
that there is no correlation with the scaling line until the
lowest doping state around $p = 1/8$ and lower.

What can we conclude from this? One implication
may be that in this heavily underdoped region the
in-plane physics is rather similar to the out-of-

plane physics, namely that of Josephson-coupled SC
domains. Here the electronic state is probably granular,
with SC patches separated by insulating Josephson bar-
rriers. Such a model has been proposed even for opti-

mally doped cuprates and STM suggests the presence of
nanoscale inhomogeneity\cite{23, 24}. However this has been
questioned\cite{27}, and in particular the electronic state seen
from the perspective of NMR and specific heat seems
to be homogeneous for $p > 0.12$\cite{28}. Below that doped
state these techniques reveal the presence of normal
quasiparticles\cite{27}. Here the electronic state definitely ap-
ppears to become spatially heterogeneous.

We turn now to ask the question as to whether the
universal scaling relation may also apply in the case of
impurity scattering. This behavior can be understood in
terms of generally-accepted theories for the pair-breaking
effects of Zn impurities, namely the suppression of $T_c$ by
non-magnetic scattering, which for a $d$-wave supercon-
ductor suppresses $T_c$ according to the Abrikosov-Gor’kov
(A-G) formula and $\rho_s$ according to the work of Maki and
coworkers\cite{28}. When we add in the effects of scattering
on $\sigma(T)$ (described by Matthiessen’s rule) this roughly
reproduces the observed scaling behavior for optimally-
doped samples but, again, we expect large deviations for
 overdoped samples for which conductivity data as a func-
tion of Zn doping are not yet available. The theory can
be developed along the following lines.

For a non-magnetic scatterer $d$-wave SC is rapidly sup-
pressed and the effects on $T_c$, $\rho_s$ and $\sigma$ as a function of
impurity concentration, $y$, are governed by the density
of states (DOS) at the Fermi level. These effects have
been shown to be fully consistent with thermodynamic
data\cite{29}. The reduction in $T_c$ is given by:

$$-\ln(T_c/T_{c0}) = \psi(1/2 + \Gamma/T_c) - \psi(1/2), \quad (3)$$

where $\psi[x]$ is the digamma function, $T_{c0} = T_c(y=0)$.
For unitary scattering, $\Gamma = n_i/\pi g(E_F)$ is the pair-breaking
scattering rate and $\Gamma_c$ is its critical value for which $T_c$
is suppressed to zero. Here $g(E_F)$ is the DOS per spin,
$n_i(= \alpha g/abc)$ is the density of impurities, $\alpha$ is the
number of CuO$_2$ planes per unit cell and $a$, $b$ and $c$
are the lattice parameters. As before\cite{29}, we adopt the
strong-coupling scenario $\Gamma_c = 1.65k_BT_{c0}$ and determine
$g(E_F)$ from the electronic specific heat coefficient, $\gamma$,
using $\gamma = (2/3)!k^2g(E_F)$. The following linearized form
of the A-G equation is valid up to about $(2/3)\Gamma_c$:

$$T_c/T_{c0} = 1 - 0.69\Gamma/T_c = 1 - (0.86\alpha R/\gamma T_{c0}) \times y, \quad (4)$$

where $R = N_Ak_B$. In the overdoped region $\gamma$ is con-
stant, independent of temperature and doping, so that the
slope, $\partial T_c/\partial y$, in eqns. (3) or (4) remains constant.
As a practical measure\cite{28}, the value of $\gamma$ was taken as
$S/T$ evaluated at $T_c$, where $S$ is the electronic entropy.
This is just the average of $\gamma(T)$ between $T = 0$ and $T_c$.

The calculation of the reduction in superfluid density
is more complex and is carried out numerically\cite{28}. How-
ever, we find an excellent approximation to within a few
percent across the entire pair-breaking range using:

$$\rho_s/\rho_{s0} \approx \frac{T_c/T_{c0}}{1 + 1.7(1 - T_c/T_{c0})}. \quad (5)$$

This non-linear relation reflects the fact that $\rho_s$ is ini-

tially suppressed much faster than $T_c$\cite{31}. Eqns. (3)
and (5) are universal relations and if impurity scatter-
ing were to have no effect on $\sigma$ then the effect of Zn
substitution would be given by the dashed A-G curves
shown in Fig. 1(a) for the optimal and most overdoped
Y$_{1-x}$Ca$_x$Ba$_2$Cu$_3$O$_{7-\delta}$.\cite{31}

In fact, the resistivity is increased by impurity scatter-
ing according to a linear Matthiessen law. The resultant
decrease in $\sigma(T)$ seems to effectively linearize the simple
A-G curve in the scaling plot. We illustrate this using the
experimental data shown by the crosses in Fig. 1. These
are data for an optimally-doped sample where the reduc-
tion in $\rho_s$ and $T_c$ is from $\mu\text{SR}$ measurements by Bernhard
et al.\cite{32}, and the $\sigma(T)$ data for Zn substitution at the
same doping state are from thin-film samples studied by
Naqib et al.\cite{16}. These trend back linearly to the origin
in reasonably good agreement with the scaling curve. We
now seek to justify this quantitatively.

The residual planar resistivity arising from impurity scatter-
ing is given by\cite{33}

$$\rho = 4(\hbar/e^2)(n_i/n)\sin^2\delta_0 \times (\delta_0), \quad (6)$$

where $\delta_0$ is the $s$-wave scattering phase shift which, in
the unitary limit, we take to be $\pi/2$. Thus the total resistivity is

$$\sigma = \frac{ne^2\tau/m^*}{1 + (4\hbar\tau/abm^*)} \times y$$

where $\tau$ is evaluated at $T_c(y)$.

There are two impurity effects to be considered. Firstly,
there is the Matthiessen term in the denominator of
eqn.(6) and, secondly, $\sigma(T)$ must be evaluated at $T_c$
which is itself reduced by scattering. This second effect
is accommodated by the fact that, in eqn. (7) below,
the only two variables in the coefficients are $\tau$ and $T_c$
and they appear as the product $\tau T_c$. Thus, provided the
material lies on the scaling curve in the absence of impurities, this product is a constant. Proceeding, the explicit variable $y$ in eqn. (6) may be eliminated by substituting from eqn. (4) to give:

$$\sigma(y)T_c(y) = \frac{\sigma(0)T_{c0} \times (T_c(y)/T_{c0})}{1 + (\beta - 1)(1 - T_c(y)/T_{c0})},$$

(7)

where $\beta = 4\hbar^2\tau_0/0.86\alpha R_{amb}^*$ and $\tau_0 = \tau$ evaluated at $T_c(y = 0)$. Taking $m^* = 2m_e$, as observed for nodal quasiparticles,[32] we have only to note that the right side of eqn.(7) is of the same form as eqn.(5) with the coefficient $(\beta - 1) = 1.9$. Thus

$$\sigma(y)T_c(y)/\rho_s(y) = \sigma(0)T_{c0}/\rho_{s0},$$

(8)

and the impurity scattering data remain on the scaling curve if, in the first instance, it lies on the curve in the absence of impurity scattering. This seems to explain the crosses shown in Fig. 1. This calculation shows that, with underdoping, the curve would steepen due to the further opening of the pseudogap (and the associated reduced entropy at $T_c$) while with overdoping it would flatten (due to the closing of the pseudogap). It would be nice to test this but we do not have combined $\rho_s(y)$ and $\sigma(y)$ data as a function of Zn concentration for heavily overdoped samples. Nevertheless, we have calculated the expected effect of Zn substitution using eqn. (7) and this is plotted in Fig. 1 by the dotted line. This shows a fairly linear behavior but which is displaced far from the “universal” scaling curve.

In conclusion, we have examined the scaling relation, $\rho_s \propto \sigma(T_c) \times T_c$, and find this is equivalent to the relation $h/\tau = 2.7 \pm 0.5 \times k_B T_c$ which could equally arise from (i) conventional dirty-limit SC, (ii) marginal-Fermi-liquid behaviour, (iii) Josephson-coupling along the c-axis of the CuO$_2$ planes, or (iv) unitary-limit impurity scattering. We conclude that the Pb and Nb samples correlate because these samples are near the dirty limit. Interestingly, the Homes scaling line correspondes to the mean free path just above $T_c$ being approximately $2 \times \xi$ (using the maximum gap $\approx 2.38 k_B T_c$ for a weak-coupling d-wave SC). Even more intriguingly, in terms of standard microscopic BCS theory,[32] it corresponds to the Gor'kov kernel having a range $\xi_G$ near $T_c$ equal to the mean free path just above $T_c$.

When we examine the doping dependence of the in-plane $\rho_s$ and $\sigma(T_c) \times T_c$ we find a total breakdown of the scaling relation across the overdoped region. This shows that the mean free path at $T_c$ is now much larger than both of the above-noted coherence lengths. This breakdown may extend into the underdoped region, allowing for the fact that the only exception is for thin-film Y$_{1-x}$Ca$_x$Ba$_2$Cu$_3$O$_{7-\delta}$. The recovery of the scaling behavior at very low doping may arise from similar physics to c-axis Josephson coupling, namely, weak-linked patches of superconducting domains in the inhomogeneous strongly-underdoped state.

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