$n_s - T_c$ Correlations in Granular Superconductors

Y. Imry,[1,*] M. Strongin,[2] and C. C. Homes[2,†]

[1]*Department of Condensed Matter Physics, Weizmann Institute of Science, Rehovot, 76100, Israel*
[2]*Condensed Matter Physics and Materials Science Department, Brookhaven National Laboratory, Upton, New York 11973, USA*

Following a short discussion of the granular model for an inhomogeneous superconductor, we review the Uemura and Homes correlations and show how both follow in two limits of a simple granular superconductor model. Definite expressions are given for the almost universal coefficients appearing in these relationships in terms of known constants.

Introduction.—The model of a granular superconductor, in which superconducting grains are coupled via Josephson tunneling, is considered [1–4]. This model is a very useful paradigm both in its own right and because it is applicable to a number of real situations. These range from man-made Josephson arrays to a variety of inhomogeneous superconductors. Without insisting on the relevance to high-T_c superconductors, one may note that the ubiquitous phenomenon of the "pseudogap" in such materials [5] finds a very natural qualitative explanation in this model. This explanation is very clear in the high intergrain resistance case (see part A below). The real transition is the phase locking of the grains while around the (higher) T_c of the grain material, each grain develops (continuously in temperature) a fluctuating order parameter, which leads to a smaller density of states near the Fermi level. Thus, the formation of the pseudogap in this model is just a crossover, whose width is determined by, and decreases with, the grain size. These grains are evidently due to regions in the system whose effective T_c is higher due to fluctuations in the doping level. The sensitivity of T_c to the doping level, compared with the appropriate interface energy, will determine whether, effectively, grains will form (described later in the discussion). The case of a d-wave superconductor, where T_c is sensitive to disorder, is immediately highlighted.

In this Letter we consider the question of the universal correlations reported experimentally between the low-temperature superfluid density n_s and the transition temperature T_c (Refs. [6–9]). Three such correlations have been reported for high-T_c superconductors [10,11], and in some cases for usual "low-T_c" ones. Two of them are different from each other while the third may be related to the second; these will be examined shortly. It is of great interest to understand the physics behind such correlations [12,13] and their respective ranges of validity [14–16]. We show that both of these clearly different correlations follow from two limits of a simple classical granular superconductor model [1–4] and derive the coefficients in terms of natural constants and the gap-to-T_c ratio for the underlying grain material. Our derivation is embarrassingly simple.

The Uemura-Homes law follows when the critical temperature for the intergrain phase locking is much smaller or comparable to that for the grain material. The strongly inhomogeneous, granular, picture is a broadly applicable paradigm [17], describing many diverse systems [3,18,19]. Recently, there is strong evidence for the relevance of this paradigm also for high temperature superconductors [20].

In 1988, Uemura *et al.* [6] reported, for underdoped high-T_c superconductors, the proportionality of n_s/m^* (or λ^{-2}, where λ is the penetration length and m^* the effective carrier mass, which is of the order of $5m$ for most of the considered materials, where m is the electron mass) to T_c. Here n_s was determined from the muon spin relaxation rate for four high T_c families with varying doping level (carrier density). The coefficient in the linear relationship is such that a carrier density of $n_s = 2 \times 10^{21}$ cm^{-3} corresponds to $T_c \simeq 25$ K.

In 2004, Homes *et al.* [7] reported a different correlation, valid more generally, including the overdoped and optimally doped cases: $\rho_{s0} \simeq 120 \sigma_{dc} T_c$, where ρ_{s0} is the strength of the condensate determined by optical measurements, and σ_{dc} is the normal-state dc conductivity near T_c. The superfluid density is related to the superconducting plasma frequency $\rho_{s0} \equiv \omega_{ps}^2 \propto n_s/m^*$ as well as the penetration depth $\rho_{s0} = c^2/\lambda_0^2$. Nine different high-$T_c$ material families with varying doping (including optimal and beyond) were examined, as well as the phonon-mediated superconductors Pb and Nb, shown in Fig. 1. This result has been interpreted [8] in terms of the conventional decrease of n_s proportional to $\ell/\xi_0 \propto T_c \tau$ in the dirty limit of BCS superconductors, where τ and ℓ are the mean-free time and scattering length and ξ_0 the zero-temperature BCS coherence length ($\xi_0 \propto v_F/T_c$). The questions of why these materials are in the dirty limit, when T_c is so high and the coherence length so small, and to what extent can the BCS-type relationships be used for high T_c materials (in spite of current theoretical beliefs) were left open. Clearly, the d-wave nature of these superconductors might play an important role here. Finally, in 2005 Zuev *et al.* [9] reported a linear relationship between n_s and T_c^χ, where $\chi = 2.3 \pm 0.4$. They pointed out that with the empirical
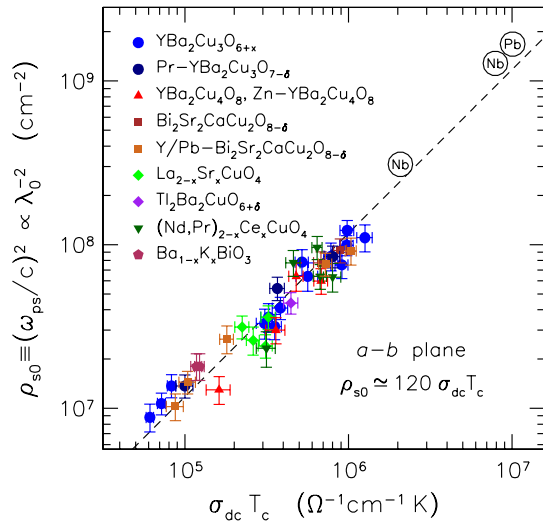
FIG. 1 (color online).   A log-log plot of the superfluid density ρ_{s0} vs $\sigma_{dc}T_c$ in the a-b planes for a variety of electron and hole-doped cuprates. The dashed line corresponds to the general result for the cuprates $\rho_{s0} \simeq 120\sigma_{dc}T_c$. The points for Nb and Pb, indicated by their atomic symbols, also fall close to this line.

proportionality of T_c to σ_{dc} (theoretically justified in a large parameter regime for a classical Josephson-coupled superconductor [2], see below), a value of $\chi = 2$ is within the experimentally determined range and would make their result consistent with the one by Homes *et al.* [7].

These quite universal correlations have caused considerable discussion [12]. For a recent explanation, we mention the one relying on the vortex glass melting temperature [13].

Method.—We shall now demonstrate that the various $n_s \propto T_c$ relations follow in an almost trivial manner for a classical (no capacitive energies) ordered Josephson array [3]. We take the simplest model of a two-dimensional (2D) array of square $L \times L$ grains of thickness d in the x-y plane, made from a superconductor with a critical temperature T_c^0. The grains are connected by flat Josephson junctions with Josephson current amplitudes I_J and Josephson energies $E_J = \hbar I_J/2e$. The 2D array can be regarded as the whole system or as one of the layers in a 3D structure. From now on we mainly consider "large grains" in the Anderson sense [21]. There, the intragrain gap is much larger than the single-particle level spacing w_L of the isolated grain. In such "large" grains, bulk superconductivity is approximately valid. The Josephson coupling can be written as [22]

$$E_J = (\pi/4)g_n\Delta, \qquad (1)$$

where g_n is the intergrain conductance measured in units of $e^2/\hbar$. We assume, for definiteness, that the size L of each superconducting unit is $\ll \lambda_0$, where λ_0 is the penetration depth of the grain material. (It is straightforward to get the result for $L \gg \lambda_0$ as well). This immediately implies that

L is much smaller than the effective penetration depth of the array; i.e., all induced fields are neglected. This can be taken as a model for a granular superconductor as long as the effects of the capacitances and the intergrain disorder, which certainly exist in real cases, are not dominant [23].

We now obtain the linear response to a small magnetic field B_z perpendicular to the array. For $\lambda_0 \gg L$ the field B_z is uniform over each grain. $\vec{B}$ is derived from a vector potential $\vec{A} = (B_z y, 0, 0)$. Note that $\nabla \cdot \vec{A} = 0$ as required for the London gauge. Thus the London equation takes the form

$$j_s = -\frac{n_s e^2}{m^* c}A. \qquad (2)$$

The flux enclosed in an $L \times y$ rectangle shared equally by two neighboring grains is $B_z L y$. Due to it, the phase difference between two superconducting blocks that are nearest neighbors in the x direction, increases with y in the manner

$$\phi(y) \simeq -2eB_z yL/\hbar c = -2eLA_x(y)/\hbar c. \qquad (3)$$

For small B, this leads to a Josephson current density [22]

$$j_{s,x}(y) = -2eI_J A_x(y)/\hbar cd. \qquad (4)$$

Equating this Josephson current to the screening current in the London equation [Eq. (2)], we find the general relation for a granular superconductor with $L \ll \lambda_0$ is similar in form to the result for an array of superconducting weak links [24]

$$n_s = \frac{4m^*}{d\hbar^2}E_J. \qquad (5)$$

This relation can be written in terms of T_c and the normal-state conductivity. Different results are obtained in the two following cases.

A. Large intergrain resistance $g_n \ll 1$.—Because the electrons are well localized in the grains, one expects the normal state of this system to be insulating when extrapolated to $T \to 0$ [3]. Here, to reach $E_J \sim T$, one needs to go to temperatures much lower than T_c^0 [see Eq. (1)]. At those temperatures E_J saturates with values of order $g_n\Delta(0)$, and T_c is given by a constant ζ of order unity times E_J, $T_c = \zeta E_J$. We have used units in which $k_B = 1$ throughout. Thus, in this case we obtain

$$n_s = \frac{4m^*}{d\hbar^2\zeta}T_c, \qquad (6)$$

which is just the Uemura correlation. For $m^* = 5m$, $\zeta = 1$, $d = 5$ Å, and $n_s = 2 \times 10^{21}$ cm^{-3}, we obtain $T_c \approx 35$ K. Thus, the coefficient in Eq. (5) agrees within a factor of two with the Uemura one, for reasonable parameters of the 2D layer. Equation (5) is just the relation between n_s and the order-parameter phase stiffness for the XY model. In this limit, there exist two distinct effects, the buildup of the pairing correlations in the grains, which is a continuous

crossover around T_c^0, and the intergrain phase locking at T_c. Were this picture applicable to the underdoped high-T_c case, T_c^0 and T_c would correspond to the establishment of the pseudogap and that of overall superconductivity respectively.

When the Uemura correlation was first reported, the proportionality of T_c to the electron density was taken to indicate the purely electronic origin of high-T_c superconductivity. Our simple derivation above proves that that logic is not infallible. The Josephson array can model any appropriately inhomogeneous superconductor, including ordinary low-T_c ones, and it does yield the Uemura correlation.

B. Small intergrain resistance $g_n \gtrsim 1$, *(including* $g_n \gg 1$).—Here E_J becomes comparable to T around T_c^0, which is then approximately equal to T_c. In the high-T_c case, this would mean that the pseudogap and the superconductivity are established at the same temperature, which is the case for the optimally doped and overdoped situations. Here we obtain, defining the constant A ($\cong 1.75$ in the BCS case) via the usual relationship in the grain material, $\Delta(0) = AT_c^0$:

$$n_s = \frac{A\pi g_n m^*}{d\hbar^2} T_c = \frac{A\pi \sigma'_n m^*}{\hbar^2} T_c, \qquad (7)$$

which is equivalent to the Homes' law. Here σ'_n is the normal state conductivity just above T_c, in units of $(e^2/\hbar)$/cm. It makes sense that the condition $g_n \gtrsim 1$ is in fact satisfied for high enough doping. While the square of the superconducting plasma frequency is defined as $\omega_{ps}^2 = n_s e^2/(\pi m^*)$ in units s^{-2}, the quantity that Homes *et al.* examine is $(\omega_{ps}/c)^2$ in units of cm^{-2}; this allows us to cast the Homes' law in the following seemingly elegant way. Using the "universal" constant $\alpha' \equiv A\alpha \simeq 0.0128$, where $\alpha \equiv e^2/(\hbar c)$ is the fine structure constant and we have previously taken $A = 1.75$

$$(\omega_{ps}/c)^2 = \pi \alpha' \sigma'_n T_c/(\hbar c). \qquad (8)$$

The approximate coefficient in the proportionality of the left-hand side to $\sigma'_n T_c$ depends only on the gap-to-T_c ratio and on natural constants. We note that this result agrees exactly with the usual "dirty limit," in which the reduction of n_s is $n_s = n(v_F \tau/\xi_0)$. Using the BCS-type relationship $\xi_0 = \hbar v_F/[\pi \Delta(0)]$ yields Eq. (8). Since the dirty-limit value agrees approximately with the Homes' law coefficient [7,8], so should our result [Eq. (8)].

In fact, since $e^2/\hbar \simeq 1/4100 \ \Omega^{-1}$, $\sigma'_n \simeq 4100\sigma_n$, when σ_n is measured in Ω^{-1}cm^{-1}. This yields

$$(\omega_{ps}/c)^2 \simeq 50\sigma_n T_c/(\hbar c) \simeq 235\sigma_n T_c. \qquad (9)$$

In the last expression on the right-hand side σ_n and T_c are in the same units as in the original Homes law paper [7] (namely, Ω^{-1}cm^{-1} and K) [25]. The slope in Eq. (9) is larger by about a factor of two from the value 120 ± 25 reported in Ref. [7]. Note that this expression can be

recast as $T_c \propto \rho_{s0}\rho_{dc}$, where $\rho_{dc} = 1/\sigma_n$, allowing a more direct comparison with the Uemura relation. For the low-T_c cases, the agreement is within about 50 percent. Taking, as an example, the first Nb sample of Table I of Ref. [8], we get from the values of σ_n and T_c, $(\omega_{ps}/c)^2 \sim 4.7 \times 10^8$ cm^{-2}, while the value in the table is $\sim 3.1 \times 10^8$ cm^{-2}. Overall, the agreement of our Eq. (9) with experiment is within a factor of two. With the gross simplifications introduced in our naïve model, we regard this as satisfactory.

Discussion.—Nominally, the high-T_c superconductors look like they are in the clean limit. This is believed rather generally and is consistent with the values reported in Refs. [7,8]. This is not because they are so clean; the experimental values of $k_F\ell$ can be of order 50–100. The problem is that ξ_0 is small due to the relatively large ratio T_c/E_F (here $k_F\xi_0 \sim 10$–50). Thus, these materials would be in the clean limit if they were homogenous. However, the natural fluctuations in the doping make them inhomogeneous, and ξ_0 is not large enough to average that. What is shown above is that this inhomogeneous system behaves like a dirty one. (Although, again, it would not be, were it homogenous.) How the inhomogeneity arises has been discussed by Alvarez and Dagotto [17] and more recently by Hoffman [26]; we elaborate on this in Ref. [3] following the general argument by Ma and one of us [27]. The issue at hand is to find the size scale where the gain in energy $\propto L^{d/2}$ due to fluctuations in the defect concentration or doping is larger than the energy of the interface created $\propto L^{d-2}$. When this scale of L is smaller than the effective correlation length, the instability and formation of grains will occur. Clearly, a stronger sensitivity of T_c to the defect concentration will help in the establishment of the granular state of matter. As mentioned before, a d-wave character of the superconductor is very helpful in that respect.

The behavior of the granular model presented is determined by two conditions, governed by dimensionless ratios: whether the grains are large or small [depending on the ratio $\Delta(0)/w_L$] and whether the intergrain resistance (in units of $\hbar/e^2$) is larger or smaller than unity. The Uemura correlation is valid only for the large grain, large resistance case which typically corresponds to the underdoped region of the phase diagram for the cuprates where a pseudogap is usually observed [5]; $\Delta(0)/w_L \gtrsim 1$, $g_n \ll 1$, where the insulator and the inhomogeneous Josephson phase are the relevant phases. In the large grain, small resistance case (which corresponds to the optimally or overdoped cuprates where the pseudogap is either strongly diminished or absent altogether), $\Delta(0)/w_L \gtrsim 1$, $g_n \gg 1$, we showed that the Homes' law is the relevant correlation. In the small grain case $\Delta(0)/w_L < 1$ and small resistance metallic regime $g_n \gtrsim 1$ (the optimal and overdoped regime), superconductivity is established in an almost homogenous, strongly disordered, conductor. Even close to the metal-insulator transition the mean free path ℓ is of a small

microscopic size and it makes sense that the superconductor should be in the dirty limit ($\ell \ll \xi_0$). This implies that the Homes' law [7,8] (or the one reported by Zuev *et al.* [9]) should then yield the valid correlation between n_s and T_c. The small grain case $\Delta(0)/w_L \lesssim 1$ with large intergrain resistance $g_n \lesssim 1$ (and therefore an insulating normal state) is very interesting since strong intragrain superconducting correlations do exist [28], but it is not known exactly what is the effect of the intergrain coupling with the Coulomb blockade [23].

We have neglected throughout the capacitive, Coulomb-blockade-type interactions [23]. This is justified in all the metallic regimes, due to screening. This includes all the range of the Homes correlations. The capacitive effects might also play an important role in the large-grain-small-intergrain-coupling case, where the Uemura correlations are supposed to hold. Our treatment there is valid only for grains large enough for the capacitive effects not to be important.

Only compact regular grains were considered in this Letter. More general inhomogeneities (e.g., stripes [11], layers, or more complex geometries) should be treated as well. The case of high-T_c materials is further complicated due to the anisotropic gap and correlation length [29]. The question of when such a superconductor can be regarded as dirty is beyond the scope of this Letter.

The results presented in this Letter are valid for granular superconductors and Josephson arrays. They do explain semiquantitatively the $n_s - T_c$ correlations in ordinary and in high-T_c superconductors. This obviously does not prove that the latter conform to the naïve model discussed. However, it might be taken as further evidence that the inhomogeneities, which do exist [20] in these materials, play a role in their fascinating physics.

Conclusions.—The examination of the model for a granular, inhomogeneous superconductor reveals that it can mimic a dirty-limit material. Scaling relations may be derived in the two limits of a simple granular superconductor model. In the large grain, high resistivity case [$\Delta(0)/\omega_L \gtrsim 1$, $g_n \ll 1$] Uemura-type $T_c \propto n_s$ (or $T_c \propto \rho_{s0}$) scaling is expected; however, in the small resistance case [$\Delta(0)/\omega_L \gtrsim 1$, $g_n \gg 1$] it is demonstrated that the $T_c \propto \rho_{s0}\rho_{dc}$ scaling described by Homes *et al.* is expected. In the small grain, small resistance case [$\Delta(0)/\omega_L < 1$, $g_n \gg 1$], $T_c \propto \rho_{s0}\rho_{dc}$ is again expected. Within the context of the high-temperature superconductors, the high-resistivity case corresponds to the underdoped (pseudogap) region of the phase diagram where Uemura-type scaling is observed while the low-resistivity case corresponds to the optimally and overdoped region where the pseudogap is largely absent and $T_c \propto \rho_{s0}\rho_{dc}$ is observed rather than $T_c \propto \rho_{s0}$.

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*yoseph.imry@weizmann.ac.il
†homes@bnl.gov

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