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An inhomogeneous Josephson phase in thin-film and high-T_c superconductors

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Abstract

In many cases inhomogeneities are known to exist near the metal (or superconductor)–insulator transition, as follows from well-known domain-wall arguments. If the conducting regions are large enough (i.e. when the $T = 0$ superconducting gap is much larger than the single-electron level spacing), and if they have superconducting correlations, it becomes energetically favorable for the system to go into a Josephson-coupled zero-resistance state before (i.e. at higher resistance than) becoming a "real" metal. We show that this is plausible by a simple comparison of the relevant coupling constants. For small grains in the above sense, the electronic grain structure is washed out by delocalization and thus becomes irrelevant. When the proposed "Josephson state" is quenched by a magnetic field, an insulating, rather than a metallic, state should appear. This has been shown [J. Tu, M. Strongin, Y. Imry, cond-mat/0405625 (2004)] to be consistent with the existing data on oxide materials as well as ultra-thin-films. We discuss the Uemura correlations versus Homes' law, and derive the former for the large-grain Josephson array (inhomogenous superconductor) model. The small-grain case behaves like a dirty homogenous metal. It should obey Homes' law provided that the system is in the dirty supeconductivity limit. A speculation as to why that is typically the case for d-wave superconductors is presented.
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1. Introduction

The intriguing connection between underdoped high-T_c superconductivity and the properties of disordered and granular superconductors [1–3] has been discussed ever since the discovery of these superconductors. We argue in this paper that near the superconductor–insulator (S–I) transition, inhomogeneities may lead to a zero-resistance Josephson-coupled state, which exists both in high-temperature superconductors and "usual" superconductors even though the interactions [4] causing the superconducting state may indeed be very different. Here we review and strengthen the arguments of Ref. [1] and discuss their relevance to the intriguing n_s–T_c correlations [5–8].

The underlying principle is that disorder implies inhomogeneities on some length-scales, as was first argued, in this context, by Kowal and Ovadyahu [9]. These scales depend on the nature and strength of the disorder. This picture is supported by numerous experiments [9,10] and may be related to domain formation by random-field-type impurities [11] (see also [12]). For example, if the Mott-type metal-insulator transition were in fact first order, as originally argued by Mott, then the arguments of Ref. [11] would imply "domain" formation in effectively 2D systems even for the weakest strength of the impurities! Finite-strength impurities will generically lead to domain formation in most situations, except very close to an appropriate second-order transition in cases where the correlation

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length diverges strongly enough [12]. Experiments considering the effect of inhomogeneities brought about by fluctuations in the local electron density or concentration gradients already exist in the literature [10]. On the theoretical side, the importance of inhomogeneities has been highlighted by Emery et al. [13], and Dagotto et al. [14]. Ghosal et al. [15] have considered a model based on the Bogoliubov-de Gennes equations, of how "homogeneous" disorder introduces an inhomogeneous pairing amplitude in ultra-thin-films. We would like to add to these interesting models that in the non-superconducting state, the phases of these domains are not locked and therefore the phase fluctuations should average the local pairing amplitude, Δ, to zero (however, $<|\Delta|^2> \neq 0$). Refs. [16,17] discuss Bose–Hubbard models and cite earlier theoretical references related to disordered systems. In recent work on ultra-thin-films [18] (see also [19]), it was argued that in an inhomogeneous medium it is possible for a Josephson-coupled superconducting state to be more stable at or near the S–I transition boundary (more disorder/less carrier density) than the metallic state (which is defined here as being on the metallic side of the percolation/localization transition). This general problem was treated some years ago in Ref. [19] using considerations based on the Thouless [20] arguments for the onset of localization in 1D and handling the Coulomb effects in the spirit of the phenomenological arguments of Abeles and Sheng [21] and Kawabata [22]. This will be reviewed in Section 2. A simple case where this clearly works is an array of Josephson-coupled clusters with an energy gap that is larger [19,23] than the energy level spacing in the cluster (see below). In this paper we are interested in extending these ideas to give some insight into weak superconductivity in inhomogeneous systems, and thus attempt to understand data in films as well as in underdoped high-T_c superconductors. In particular, we will present some insights on the Uemura correlation [5] and "Homes' law" [6,7] (see also [8]).

2. Scales for inhomogenous and granular systems

Here we briefly describe a simple argument which indicates that in an inhomogeneous system there may be a regime in which an inhomogenous Josephson-coupled state occurs before the metallic state, as the sample resistance decreases from a resistance characteristic of the insulating state to that of a conducting one. This is done by either increasing the doping in the high-T_c case, or changing the thickness in the ultra-thin-film case.

Without interactions, the Thouless picture of localization in 1D can be generalized to analyze the electronic couplings between "metallic regions" [20] in an inhomogeneous system (which can consist of grains or doped regions with high conductivity) in any dimension [24,25]. The intergrain coupling energy is given by $\hbar/\tau_L = V_L$ where τ_L is the lifetime for an electron in one of the conducting regions, of linear size L, to go into the next one. The conductance between "grains" can be related to the ratio of this coupling energy to the energy level spacing in the grains and is written as a dimensionless conductance, $g_L = V_L/w_L = 2\hbar/e^2R_L$, where w_L is the characteristic energy level spacing in the small metallic regions. When the typical intergrain resistance $R_L > 2\hbar/e^2$, then the noninteracting system becomes localized.

We now introduce the simplified Coulomb interaction, parametrized by a single capacitive energy (see below). We start with the analysis of Abeles and Sheng [21] to estimate the resistance between isolated grains where the coupling energy overcomes the intergranular Coulomb energy of $E_{coul} = e^2/2C_L$. By approximating $\hbar/\tau_L$ as $\hbar/R_LC_L$, and setting this equal to $e^2/2C_L$, one gets the same value as before, of $R_L \simeq 2\hbar/e^2$ for the resistance below which the "intergranular" coupling is greater than the Coulomb repulsion (where R_L is the tunneling resistance between grains and C_L is the mutual capacity of the two grains). Thus in this case a system with Coulomb interactions will also be metallic once $R_L < 2\hbar/e^2$. Clearly, these two approaches are not unrelated. A physical argument relating them might be based on the fact that once the single-electron eigenfunctions are delocalized and spill over from the grain, the Coulomb blockade picture with quantized charge on the grain becomes meaningless. Evidently, this argument is certainly valid when the Coulomb energy is weak, $e^2/2C_L < w_L$, and it is treated as a perturbation on the noninteracting picture. The argument may also hold for strong interactions, $e^2/2C_L > w_L$, provided that the actual value for R_L, which may be strongly renormalized by the interaction, is used. In Ref. [19] the noninteracting picture was generalized, following Ref. [22], to include the effect of strong Coulomb interactions (i.e. $e^2/2C_L > w_L$), which of course is typically crucial due to the marginal screening and the charging energy when electrons move between conducting regions. Here, to get metallic behavior, the intergrain transfer energy should overweigh the Coulomb energy [19,22] (see also [24,25])

$$zV_L = 2\hbar w_L z/e^2R_L > e^2/2C_L, \tag{1}$$

where z is the coordination number, related to the typical number of nearest neighbors, which appears in the mean-field theory for the transition.

The condition for superconductivity is, however, that the Josephson energy, given by the standard expression $E_J = \pi\hbar\Delta(0)/4e^2R_L$, be larger than the Coulomb energy, or, including again the factor z for a medium composed of grains we replace E_J by $zE_J \sim zV_L\Delta\ (0)/w_L$

$$z\pi\hbar\Delta(0)/4e^2R_L > e^2/2C_L. \tag{2}$$

Here $\Delta(0)$ is the gap at $T = 0$. In other words, the pair transfer matrix element E_J replaces here the single-electron coupling energy V_L. For this approximate argument at low temperatures, there is no need to put in the temperature dependence of the gap. The interesting consequence is that there clearly exists an unusual regime where E_{coul} can be greater than zV_L, but less than E_J, as long as $\Delta(0)/w_L$ is greater than one. So for grains that are "large" in the sense

[23] that $\Delta(0)/w_L \gg 1$, superconductivity is easier to achieve than normal conductivity [1,19,24]. This argument is only meant to show that if there are intrinsic inhomogeneities and the system has superconducting regions, then it is possible to have a Josephson state before having a metallic one.

A possible phase diagram, for $\Delta(0) \gg w_L$, is described in Fig. 1. It can be seen that at low temperatures as the conductivity increases, (by increasing the thickness in the case of films and increasing the doping in underdoped high-T_c's) one first goes from the insulating phase into the Josephson phase (line A–B) and finally into the true metallic/superconducting phase where the respective single-particle "wavefunctions" become delocalized [1,19,26] (Note that line C–B, and its continuation to higher temperatures, eventually becomes a smooth crossover rather than a sharp transition. We do not know whether the change from the Josephson phase to the usual superconductor is affected by a real transition). This is consistent with the above argument showing that if there are superconducting correlations in the insulating regime, then the quantum transition to a Josephson state can occur (for large "grains" where $\Delta(0)/w_L > 1$) before the percolation–delocalization transition.

What happens for small grains, $\Delta(0)/w_L \ll 1$? Here, by decreasing disorder, one goes from the insulator first into the metallic, delocalized phase. In this metallic phase the single-electron wavefunctions are no longer localized in the grains. The granular inhomogeneity is thus not effective for the electrons! Therefore, by decreasing the disorder further, or increasing the interactions responsible for superconductivity, the system will go into the superconducting state as a continuous (not a granular!) system.

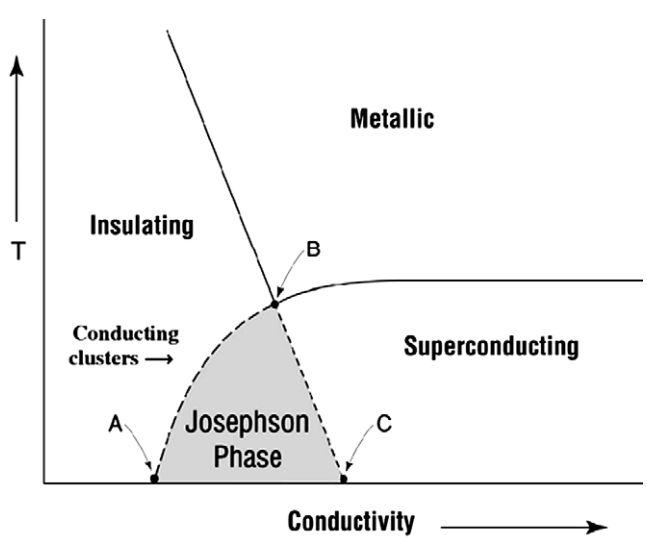

Fig. 1. The schematic "phase diagram" of an inhomogeneous "large-grain" ($\Delta(0)/w_L \gg 1$) superconductor in the temperature/conductivity plane. The conductivity is used as a measure of disorder and increases with doping or film thickness. The A–B line is the boundary between the insulating phase and the Josephson-coupled state where the superconducting regions are phase coherent. The B–C line is where the system goes into a delocalized bulk superconducting phase. In this region to the right of line B–C we would expect normal metallic conduction when superconductivity is quenched by a magnetic field. In the Josephson phase a logarithmic behavior in the resistivity seems common when superconductivity is quenched by a field. The note in the figure indicates a region to the left of line A–B where there exist disconnected metallic regions with nonzero $<|\Delta|^2>$ at low temperatures. Likewise, disconnected insulating regions may occur to the right of line A–B.

Does experimental evidence exist for our conjectures? Some of this evidence, in both ultra-thin-films and underdoped high-temperature superconductors, has been discussed in Ref. [1]. In Section 3, we review the correlations (discovered experimentally) between the superfluid density n_s and the transition temperature T_c and how they can be easily understood from our picture. Before doing that, we remark that the insulating state with intragrain superconducting correlations and $<|\Delta|^2> \neq 0$, but without intergrain phase locking, provides a simple example where a local "pseudogap" may exist above T_c. This pseudogap will have, with respect to the local "crystal" axes, the same symmetry and nature as the superconducting gap below T_c. This agrees with recent angle-resolved photoemission studies [27].

3. The n_s–T_c correlations

Here we consider the question of the universal correlations reported experimentally between the low-temperature superfluid density n_s and the transition temperature T_c. Three such correlations have been reported, for underdoped high-T_c superconductors 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 (see below). It is of great interest to understand the physics behind such correlations and their respective ranges of validity.

In 1989, Uemura et al. [5] reported the proportionality of n_s/m^* (or λ^{-2}, where λ is the penetration length and m^* the carrier mass, which is of the order of $5m$ for the considered materials) to T_c; 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 $2 \times 10^{21}/\text{cm}^3$ corresponds to $T_c \simeq 25$ K.

In 2004, Homes et al. [6] reported a different correlation: $N_c \simeq 4.4\sigma_{dc}T_c$, where $N_c = \rho_s/8$ is the spectral weight, determined by optical measurements, associated with the superconducting condensate and σ_{dc} is the normal-state dc conductivity near T_c. Nine different high-T_c material families with varying doping (including optimal and beyond) were examined, as well as the usual superconductors Pb and Nb. This result has been interpreted [7] 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 to what extent the BCS-type relationships can 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. [8] 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 proportionality of T_c–σ_{dc} (theoretically justified in a classical Josephson-coupled superconductor [19]), the value of 2 for χ makes their result consistent with the one by Homes et al. [6]. Obviously $\chi = 2$ is well within the experimentally determined range.

We shall now demonstrate that the $n_s \propto T_c$ relation holds for a classical ordered Josephson array, when the size, L, of each superconducting unit is $\ll\lambda$. This can be taken as a model for a granular superconductor as long as the effect of intergrain disorder, which certainly exists in real cases, is not dominant.

Consider for simplicity a regular infinite 2D array of square superconducting grains of linear size L and thickness d, connected by flat Josephson junctions with Josephson current amplitudes I_J and Josephson energies $E_J = \hbar I_J/2e$. The generalization to 3D is straightforward. We obtain the linear response to a small magnetic field B perpendicular to the array. For $\lambda \gg L$ the field B is uniform over each grain. B is derived from a vector potential $\vec{A} = (By, 0, 0)$. Note that $\operatorname{div} A = 0$ as required for the London gauge. Thus the London equation takes the form

$$j_s = -\frac{n_s e^2}{m^* c} A. \tag{3}$$

Due to the flux, the phase difference between two superconducting blocks that are nearest neighbors in the x direction, increases with y in the manner

$$\phi(y) \simeq -2eByL/\hbar = -2eLA_x(y)/\hbar. \tag{4}$$

The Josephson current density [28]

$$j_{s,x}(y) = -2eI_J A_x(y)/\hbar, \tag{5}$$

follows for $B \to 0$. Comparing with the London Eq. (3), we find the Uemura-type relation

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

where the constant of order unity ζ is defined via $T_c = \zeta E_J$, and we have used units in which $k_B = 1$ throughout. 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, this n_s/T_c ratio agrees within a factor of two with the Uemura one, for reasonable parameters of the 2D layer. Eq. (6) is just the relation between n_s and the order-parameter phase stiffness for the x–y model.

When the Uemura correlation was first reported, the proportionality of T_c to the 2D electron density was taken to indicate the purely electronic origin of high-T_c superconductivity. Our simple derivation above, proves that the logic is not infallible. The Josephson array can model any appropriately inhomogenous superconductor, including ordinary low-T_c ones, and it does yield the Uemura correlation.

The Uemura correlation should thus be valid for the large grains, $\Delta(0)/w_L > 1$ case, where the inhomogenous Josephson phase is the relevant one. In the small-grain case $\Delta(0)/w_L < 1$, superconductivity is established in a homogenous, strongly disordered, conductor. Close to the metal-insulator transition the mean free path ℓ is of a small microscopic magnitude and it makes sense that the superconductor should be in the dirty limit ($\ell \ll \xi_0$). This implies [7] that Homes' law [6,7] (or the one reported by Zuev et al. [8]) should then yield the valid correlation between n_s and T_c. The case of high-T_c materials is further complicated due to the anisotropic gap and correlation length. The question of when can such a superconductor be regarded as dirty is interesting and nontrivial. Its full analysis is beyond the scope of this paper. We speculate in the next section that the fact that in the nodal regions $\Delta(0)\tau \ll 1$ even for weak disorder, may well be relevant.

4. Some experiments on inhomogenous superconductors and thoughts on high-T_c

In this short section we would like to discuss how the inhomogeneities we have previously mentioned can be consistent with the correlations found by Homes et al. [6,7]. These issues involving inhomogeneities occur not only in the high-T_c superconductors, but also in some other granular systems. For example we briefly mention that both underdoped high-T_c superconductors [29] and other systems such as NbN [30] show a logarithmic dependence on temperature for the resistance in a magnetic field. Beloborodov et al. [31] have mentioned that this logarithmic behavior can occur in granular systems and high-T_c superconductors. Furthermore, tunneling microscopy (although not completely understood) also shows evidence that high-T_c superconductors are not physically homogeneous [10]. We have previously mentioned that there are two limits for inhomogenieties in superconductors. Those are the Josephson phase, where $\Delta(0)/w_L \gg 1$, and the small-grain case where $\Delta(0)/w_L \ll 1$ and the various regions of the film are connected.

A summary of the nature of the films that satisfy these conditions has been given some time ago [1,19]. The small-grain case is relatively simple and will not be further discussed in this paper. In the high-T_c superconductors the situation is more complex and in the rest of this section we would like to deal with Homes' law [6,7] and why it works there. Tunneling microscopy [10] seems to indicate that high-temperature superconductors are consistent with the condition that $\Delta(0)/w_L > 1$. We have already indicated that Homes' law seems consistent with a dirty superconductor and in fact it works for dirty conventional metallic films as well as high-T_c superconductors. The question is why does this apply for high-temperature superconductors, which microscopy shows are granular in nature. Granular systems where $\Delta(0)/w_L \gg 1$ can indeed be superconducting, but they do not necessarily act like dirty limit superconductors in the Ginzburg–Landau theory, unless the coherence

length is long. It is possible that the d-wave nature in high-temperature superconductors provides a way for this to happen. In the nodal region the superconducting gap is small and the coherence length may be large. These considerations are discussed in an interesting paper by Joglekar et al. [32]. This situation, where there is a large coherence length (in the nodal region) compared to the mean free path, may bring the system back to the dirty limit and this could serve as a possible way to understand Homes' law in high-T_c superconductors.

5. Conclusions

We proposed the picture of spontaneously formed conducting domains which form a Josephson phase at low temperatures, as a general description for some disordered systems near the superconductor–insulator transition, especially in the effectively 2D case [11]. As far as we know, there is really no experimental evidence for a uniform state at the S–I transition. In this regime where large superconducting regions first appear (to the left of line A–B in Fig. 1) they are initially decoupled (this region which is analogous to the pseudogap state is not shown). As line A–B is approached, Josephson coupling produces phase alignment of the order-parameter of different superconducting regions, and this happens before the percolation–delocalization transition to the metallic state, along line C–B. We discussed the various n_s–T_c correlations and showed how the Uemura correlation naturally arises for the Josephson array superconductor. The Homes correlations follow for a dirty superconductor and we speculated how this can arise in the d-wave case.

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