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Negative local superfluid densities: The difference between dirty superconductors and dirty Bose liquids

B. I. Spivak and S. A. Kivelson

Department of Physics, The University of California at Los Angeles, Los Angeles, California 90024

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We show that in a disordered superconductor near the superconductor-to-insulator transition, the local superfluid density N_s fluctuates from point to point in sign as well as magnitude. We demonstrate this explicitly with a simple model in which correlation effects produce a negative Josephson coupling between two superconducting grains. We argue more generally that both correlation effects and resistance fluctuations will produce random signs of N_s. This implies that a disordered superconductor is more like a quantum XY spin glass than a disordered Bose liquid.

INTRODUCTION

The nature of the superconductor-to-insulator transition has been studied extensively for many years.[1-7] It has been argued that the transition can be described in the context of the superfluid-to-insulator transition in a charged, disordered Bose liquid.[5,7,8]

Experimentally, there is an apparent distinction between granular[1,2] and homogeneously disordered[3,4] materials: In the granular materials there is reasonable evidence that a gap opens up in the quasiparticle spectrum at the bulk T_{c0} when a local order parameter develops on each grain, although the actual superconducting T_c can be much smaller than T_{c0}, or can even be 0, depending on the magnitude of the normal-state resistance; in these materials it is clearly reasonable[5,6] to imagine that at temperatures less than T_{c0}, the only important long-wavelength, low-energy physics is captured by a model in which the only relevant dynamical variable is the phase of the order parameter on the grains (with possible renormalizations of the effective parameters due to quasiparticle excitations across the junctions[5(a),9]). By contrast, in the homogeneous materials,[3,4] there is no sign of any remnant of the bulk T_{c0} in high-resistance samples, and there is good evidence[3] that as the normal-state resistance is increased, both T_c and the quasiparticle gap Δ vanish together in such a way that $2\Delta/kT_c = 3.5$; *a priori* it seems unlikely that the superconducting transition in these materials can be described in terms of fluctuations of the phase of the order parameter alone.

In this paper we shall ignore the complications due to quasiparticle excitations discussed above (e.g., imagine we are considering a granular material), and shall treat the phase fluctuations in terms of the well-known effective Hamiltonian

$$H^{\text{eff}} = -2e^2 \sum_{i,j} n_i (C^{-1})_{ij} n_j + \sum_j \mu_j n_j - \sum_{i,j} J_{ij} \cos(\theta_i - \theta_j) , \quad (1)$$

where n and θ are canonically conjugate variables,[10] $[n_j, e^{j\theta_k}] = \delta_{jk} e^{i\theta_k}$, C_{ij} is the capacitance matrix, μ_j is the site energy on grain j, and J_{ij} is the Josephson coupling between grain i and j. Here, n_j is the number of Cooper pairs on grain j. In all previous discussions,[5-8] J_{ij} was assumed to be positive. Since it is a fundamental feature of a Bose liquid in the absence of a magnetic field that the kinetic energy is unfrustrated (i.e., the ground state is nodeless), it is necessary that each J_{ij} be positive if Eq. (1) is to be interpreted as a model of a Bose system. Specifically, since $J_{ij} \sin(\theta_i - \theta_j)$ is the Josephson current across the junction between grains i and j, we recognize that J_{ij} is the lattice version of the local superfluid density $N_s(\mathbf{r})$, i.e., the proportionality constant between the supercurrent and the superfluid velocity,

$$\mathbf{J}_s(\mathbf{r}) = eN_s(\mathbf{r})\mathbf{v}_s(\mathbf{r}) . \quad (2)$$

Our aim here is to show that in a disordered superconductor, it is possible that $N_s(\mathbf{r})$ (or, equivalently, J_{ij}) can be negative. In this sense, the disordered superconductor is unlike a Bose liquid. The idea that $N_s(\mathbf{r})$ can be negative has been discussed previously.[11] However, here we present a clear demonstration that this behavior does, in fact, occur in equilibrium, even in the absence of magnetic order.

SOLVABLE MODEL

Consider a simple solvable case in which the phase description in Eq. (1) is clearly applicable, namely the case of two Josephson-coupled grains. In the usual case, in which direct single-electron tunneling between grains is responsible for the Josephson coupling, J_{12} is guaranteed to be positive in the absence of spin-orbit coupling. However, the situation is somewhat richer if we consider the case in which the tunneling is indirect, through a localized state between the two grains, as shown in Fig. 1(a). We consider the simple model problem

$$H = H_1 + H_2 + \sum_{j=1,2} \sum_{k,s} T_{jk} (c^\dagger_{jks} c_{0s} + \text{H.c.}) + \varepsilon_0 n_0 + U(n_0)^2 , \quad (3)$$

where H_j is the Hamiltonian of grain j, c_{jks} annihilates an electron with spin s and other quantum numbers k on grain j, c_{0s} annihilates an electron of spin s in the local-

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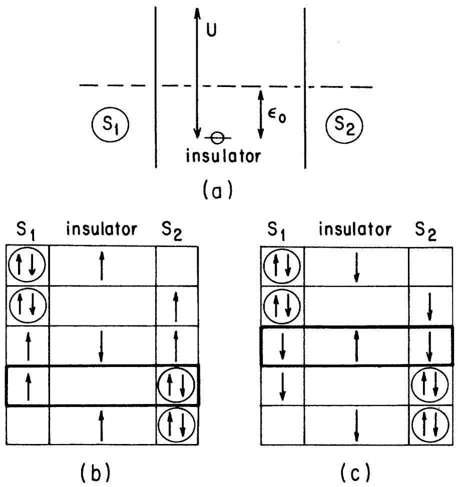

FIG. 1. (a) Schematic picture of the system modeled by the Hamiltonian in Eq. (3). (b) Schematic picture of the sequence of intermediate states that leads to the expression from fourth-order perturbation theory for the Josephson coupling in Eq. (4) in the case in which the impurity spin is up. The first box represents the initial state and the pair of electrons in a circle is a Cooper pair on grain 1, i.e., two electrons which are a part of the superconducting condensate. The first intermediate state is reached when the electron is transferred from the impurity to grain 2. Next an electron with spin down is ripped from the condensate on grain 1, and transferred to the impurity, leaving behind a spin-up quasiparticle in grain 1. It must be a spin-down electron that is transferred since it must ultimately form a Cooper pair with the initial electron that was transferred to grain 2. Indeed, next, the spin-down electron is transferred to grain 2 and forms a Cooper pair with the spin-up electron that is already there. In order that the electrons be in the cannonical order, it is necessary in this step to permute the order of the two electrons that have been transferred to grain 2. This exchange is responsible for the negative sign, hence we have enclosed this state with a heavy box. Finally, the remaining unpaired electron is transferred from grain 1 to the impurity, restoring the impurity to its original state. (c) Same as (b), except this time for the case in which the impurity spin is down. Again, the boxed step involves the interchange of order of two electrons.

ized state, and $n_0 = 0$, 1, or 2 is the number of electrons in this localized state. T_{jk} is the hopping matrix element, which we will treat as a small perturbation, ε_0 is the energy of the localized state, and U is an interaction energy, which we assume is very large. When the unperturbed ground state is such that the localized state is singly occupied, i.e., when $\varepsilon_0 < 0$ and $U + 2\varepsilon_0 > 0$, then the Josephson coupling J can be easily computed to lowest order in $|T_{jk}|$, with the result that it is negative. For simplicity, we write the expression for J in the limit $U + 2\varepsilon_0 \gg -\varepsilon_0 > 0$, and zero temperature

$$J = -\sum_{k,q} (T_{1k} T_{2q})^2 [(u_{1k} v_{2q})^2 + (u_{2q} v_{1k})^2] \times \left[\frac{1}{(\varepsilon_{1k} - \varepsilon_0)(\varepsilon_{2q} - \varepsilon_0)(\varepsilon_{1k} + \varepsilon_{2q})} \right], \tag{4}$$

where

$$u_{jq} = \left[\frac{\varepsilon_{jq} + \zeta_{jq}}{2\varepsilon_{jq}} \right]^{1/2} \quad \text{and} \quad v_{jq} = \left[\frac{\varepsilon_{jq} - \zeta_{jq}}{2\varepsilon_{jq}} \right]^{1/2} \tag{5}$$

are the usual BCS coherence factors for state q on grain j, and $\varepsilon_{jq} = (\zeta_{jq}^2 + \Delta_j^2)^{1/2}$ and ζ_{jq} are, respectively, the corresponding quasiparticle energies in the superconducting and normal states, and Δ_j is the superconducting gap on grain j.

The important point is the negative sign of J; it follows directly from the anticommutation rules of the fermion creation operators. To see this, consider the BCS wave function for one of the grains,

$$|\psi_j\rangle = \prod_q (u_q + e^{i\theta_j} v_q c_{jq\uparrow}^\dagger c_{j-q\downarrow}^\dagger)|0\rangle . \tag{6}$$

It is necessary to define a phase convention (i.e., the order of the spin-up and spin-down creation operators) which is arbitrary but being once defined must always be maintained; we have chosen to put the up spin to the left of the down spin. In the case of direct tunneling between grains, the transfer of a pair from one grain to the other preserves the spin ordering. However, in the case of indirect tunneling, due to the prohibition against double occupancy of the impurity state, the ordering necessarily gets permuted, as can be seen in the sequence of intermediate states pictured in Figs. 1(b) and 1(c). Note that this result applies independently of the initial orientation of the impurity spin.[12]

It is important to stress that the sign of J is not sensitive to details of the model, as it is based only on the existence of strong correlation which leads to the prohibition against double occupancy of the impurity state. It does not depend on the existence of a static spin moment in the localized state: Since the result in Eq. (4) is independent of the orientation of the impurity spin, thermal averaging over spin orientations has no effect on J at all. Quantum fluctuations also have little effect. For instance, consider a simple generalization of the model in Eq. (1) in which the impurity spin is antiferromagnetically coupled to another spin α,

$$H' = H + I\mathbf{S}_\alpha \cdot \sum_{s,s'} c_{0s}^\dagger \boldsymbol{\sigma}_{ss'} c_{0s'} , \tag{7}$$

so that they form a singlet in the unperturbed ground state. When we repeat the perturbative evaluation of J as in Eq. (4), the result is

$$J = -\sum_{k,q} (T_{1k} T_{2q})^2 [(u_{1k} v_{2q})^2 + (u_{2q} v_{1k})^2] \times \left[\frac{1}{(\varepsilon_{1k} - \varepsilon_0 + I)(\varepsilon_{2q} - \varepsilon_0 + I)(\varepsilon_{1k} + \varepsilon_{2q} + I)} \right]. \tag{8}$$

Note that so long as I is small compared to $|\varepsilon_0|$, this result differs only to order $I\tau/h$ from the result in Eq. (4), where $h/\tau \sim \min(|\varepsilon_0|, \Delta_1, \Delta_2)$. The physical meaning of this is transparent: One should interpret τ as the tunneling time, i.e., the typical duration of the tunneling process in imaginary time. So long as $I \ll h/\tau$, the impurity spin does not fluctuate substantially during the imaginary time interval over which the tunneling occurs. This is why the result in Eq. (8) is essentially the same as that in Eq. (4). The tunneling Hamiltonians in Eqs. (1) and (7) are derived in the context of an approximation in which the tunneling time between a grain and the impurity is zero, so the total tunneling time is determined entirely by the time the impurity spends in an excited state. For a more general treatment, the tunneling time will include a contribution from the tunneling events between the impurity and the grains. We expect that more generally Eq. (4) will be little changed so long as the spin fluctuation rate is slow compared to τ. However, even if the fluctuation rate is fast compared to τ, we simply expect a change in the magnitude of J, not its sign. For instance, J in Eq. (8) remains negative even for $I \gg |\varepsilon_0|$.

MESOSCOPIC FLUCTUATIONS

Another effect that is likely to produce a locally negative superfluid density in systems near their insulating state is related to purely one-electron resistance fluctuations. For $k_F l \gg 1$, the fluctuations in the average superfluid density have been computed using standard diagrammatic techniques.[13] The result is

$$\frac{\langle (\delta N_s)^2 \rangle}{\langle \delta N_s \rangle^2} \sim \frac{e^4}{\hbar^2 \langle \sigma_\xi \rangle^2}, \tag{9}$$

where σ_ξ is the conductance of a normal-metal sample the size of the superconducting coherence length ξ. For small $k_F l$, the fluctuations are small, but they become of order 1 as $k_F l$ approaches 1. This is certainly highly suggestive, although not a proof, that the superfluid density has a high probability of taking on locally negative values.

IMPLICATIONS

We have shown that in disordered superconductors there will occur regions with locally negative superfluid density. There are a number of interesting implications that follow directly from this observation. (i) In a small superconducting ring, if there is a segment with negative superfluid density (e.g., a Josephson junction between two pieces of the ring with a negative coupling), the ground state of the ring will spontaneously break time-reversal symmetry. The ground state will have nonzero supercurrent and magnetic flux. The ground state will thus be twofold degenerate, reflecting the broken time-reversal symmetry. Of course, at longer times, this symmetry will be restored due to thermal activation or macroscopic quantum tunneling[14] between the two states, but for all except the most minute rings, these rates can be made so slow as to be experimentally irrelevant. We would expect that for dirty metal rings with conductance of order e^2/h, there is a roughly 50% chance that the ground state will break time-reversal symmetry. (ii) For disordered granular superconductors, in which a superconducting gap opens up at a temperature well above T_c so that phase fluctuations are the only relevant excitations, the low-energy properties of the system should be describable by the Hamiltonian in Eq. (1), but with random signs of the J's. Far from the superconductor-to-insulator transition, we expect that the concentration of negative J_{ij}'s will be small, but near the transition, of order 50% of the J_{ij}'s will be negative. (iii) The Hamiltonian in Eq. (1) has been studied extensively as a model of the superconductor-to-insulator transition. In the absence of disorder and for $J > 0$, a Bose-liquid Mott transition occurs as a function of the magnitude of the capacitance.[5] Another mechanism that has been considered[7,8] is a transition for random μ_j as a function of the strength of the disorder for large enough C and $J > 0$; this is analogous to an Anderson transition in a Bose liquid. Our previous discussion implies that the random sign of J is an unavoidable feature of disordered superconductors. That negative J's are a relevant perturbation, at least in two dimensions, can be verified by noting[15] that the long-range order of the ground state of the classical XY model is destroyed by even an arbitrarily small concentration of negative J's. Thus, we feel that to understand the superconductor-to-insulator transition in granular superconductors, it is necessary to study the problem of the quantum XY spin glass. This is a fascinating problem about which very little is known. However, two common features[16] of a spin glass which may have important experimentally accessible ramifications are (a) broken time-reversal symmetry in the ground state (i.e., the existence of random, trapped fluxes in the ground state) and (b) long-time tails in the dynamics of the system associated with a broad distribution of relaxation rates.

DISCUSSION

We would like to conclude with a few qualitative remarks. First, we note that in any highly correlated system with a superconducting ground state, the present effects are likely to be particularly pronounced. In particular, it is generally accepted that the high-temperature superconductors are characterized by a large Hubbard U. Thus, whatever the nature of the ground state in the absence of disorder, we expect disorder will produce quantum XY spin-glass-type behavior in the ground state. In particular, this means that in the presence of disorder there may be no way to absolutely distinguish between an anyon superconductor[17] and a conventional superconductor. Second, we feel that the superconductor-to-insulator transition in homogeneously disordered superconductors is likely to have additional features which distinguish it from the Bose liquid. In particular, as mentioned before, there is experimental evidence[3] that the gap-to-quasiparticle (fermion) excitations vanishes at T_c, even as T_c tends to zero. Of course, these fermions can be integrated out, leaving an effective interaction between phase fluctuations which is nonlocal in time. Such interactions can change the nature of the transition entirely.[6]

Note added. After the completion of this paper, we found that a sign anomaly was noted by Glazman and Matveev[18] in the course of a study of the effect of a Kondo impurity on the magnitude of the Josephson coupling.

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