Global Phase Coherence in Two-Dimensional Granular Superconductors

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Experimental evidence is presented from studies of the onset of superconductivity in ultrathin Sn films which implies that the sheet resistance is the only relevant variable in determining the onset of global phase coherence. This result is also found by a theoretical argument involving both the phase-number and energy-time uncertainty relations.

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Granular superconducting films are usually modeled as two-dimensional arrays of Josephson junctions.[1] Their phase transition is described as a topological or Kosterlitz-Thouless-Berezinskii transition.[2] Anderson[3] and Abeles[4] introduced the central concept of a maximum normal-state coupling resistance R_J. If $R > R_J$, the electrostatic charging energy exceeds the Josephson coupling energy. This argument gives a capacitance-dependent condition for global phase coherence, and predicts that the normal-resistance threshold, above which a film loses phase coherence, should depend rather critically on geometry.

We reconsider here the normal-resistance threshold for a transition of a two-dimensional granular film to zero resistance at low temperatures, i.e., the threshold for global phase coherence.[5] Examination of the superconducting properties of ultrathin quench-deposited tin films suggests that there may be a universal resistance above which global phase coherence cannot be established. Results on the depression of T_c with increasing resistance have been reported in a number of other ultrathin film systems[6–9] and are consistent with this hypothesis.

We present a semiquantitative theoretical argument for a universal threshold independent of localization theory. It comes from the requirement that global phase coherence can only occur when both the phase-number and the energy-time uncertainty relations are satisfied. We first study a single junction using the resistively-shunted-junction (RSJ) model[10] to find a threshold criterion depending only on the resistance. The conclusions follow from the macroscopic quantum-mechanical nature of the variables characterizing the Josephson coupling.[11,12] This single-junction result is then generalized to a random two-dimensional array.

Tin films were evaporated by use of a molecular-beam vapor source through a mask onto glazed alumina substrates at 18 K in a vacuum in the 10^{-10}- to 10^{-9}-Torr range with an apparatus described elsewhere.[13] Successive increments of Sn could be added over the same area increasing the nominal thickness of a film at a given time by as little as 0.1 Å. In this way we could observe the onset of superconductivity as the thickness of a film increased and its resistance decreased.

In the inset of Fig. 1 we show $R_\square(T)$ before and after addition of only 0.75 Å of Sn which brought a decrease in the normal-state sheet resistance from 10 to 3.6 kΩ. At 10 kΩ the sample did not attain zero resistance at low temperatures, even though its resistance fell by 2.5 orders of magnitude near the bulk transition temperature of Sn. On the other hand, the 3.6-kΩ sample was a superconductor at low temperatures. These curves are an example of the difference between samples which are resistive at low temperatures (i.e., exhibit only local phase coherence), and those which exhibit zero resistance (i.e., global phase coherence). In the main part of Fig. 1 we display the very rapid onset of global superconductivity with increasing thickness for seven separate series of evaporations onto different substrates. Each nearly vertical

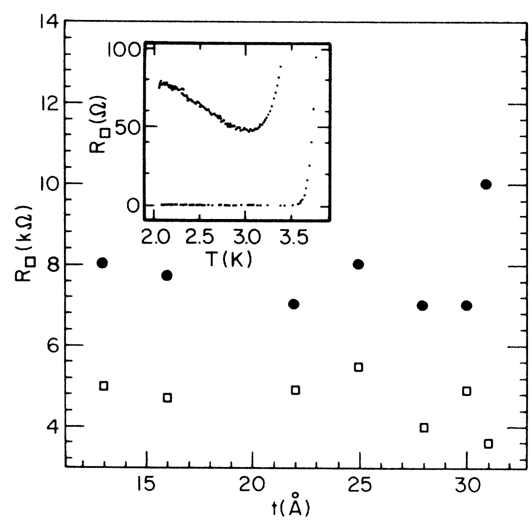

FIG. 1. Values of $R_\square(T=20\text{ K})$ vs thickness t. Each nearly vertical pair of points corresponds to successive evaporations which delineate the onset of superconductivity in seven *different* samples. When $R_\square$ has the greater value (circles), the particular sample is not a superconductor whereas after a fraction of an angstrom of Sn is added the sample becomes a superconductor with $R_\square$ given by the lesser value (squares). The inset shows $R_\square(T)$ for one of the samples.

pair of points relates to two successive evaporations of a particular series. When the film has the larger normal-state resistance (circle), it does not become a superconductor while after the next evaporation (square), it does. In each case, the nominal thickness difference between successive evaporations is a fraction of an angstrom. The pairs of points bracket a sheet resistance of roughly $1.5\hbar/e^2$. Remarkably, this resistance is about the same for each of the series of evaporations, although the nominal thicknesses vary by a factor of 3. These variations in thickness at the threshold imply significant differences in microstructure. Films with normal-state resistances higher than those represented in Fig. 1 exhibit local superconductivity: Although they do not enter a zero-resistance state their electrical resistances decrease substantially at temperatures very close to the transition temperature of bulk Sn. The behavior of these high-resistance films appears to be explained by a percolation model involving tunneling links.[13] The absence of a depression of T_c with increasing sheet resistance is in contrast with the work in Refs. 6–9. The present data together with those in Refs. 6–9 strongly suggest the existence of a universal resistance threshold below which global phase coherence is established.

A criterion for global phase coherence is found by requiring that a coupled superconducting system satisfy both the phase-number and energy-time uncertainty relations. When these fail, we can no longer describe the granular system as the quasiclassical limit of an underlying macroscopic quantum-mechanical model. One might then expect phase correlation to exist only over short distances and for short times. This can lead to a low but nonzero resistance. Our new condition for the onset of global phase coherence replaces the Anderson-Abeles requirement that the Josephson coupling energy exceed the charging energy.

To develop the argument we first treat the case of a single Josephson junction and then generalize the result to an array of junctions. Let $\Delta\phi$ and Δn be the phase-difference and pair-number–difference uncertainties, respectively.[3] They must satisfy an uncertainty relation:

$$\Delta\phi\Delta n \gtrsim \tfrac{1}{2}. \qquad (1)$$

This condition is valid provided $\Delta n \gtrsim 1$, for then $\Delta\phi \lesssim 1$ and the multiple-valuedness of ϕ is unimportant. We describe the junction by the RSJ model[10]—a parallel combination of a capacitor C, a quasiparticle resistance R, and an effective inductance $L = [(2e/\hbar)I_1 \cos\phi]^{-1}$, where I_1 is the maximum Josephson current.

In the above quasiclassical model the low-lying excitations of the junction are the plasma oscillations at the frequency $\omega_0 = 1/(LC)^{1/2}$. It is necessary to take $\hbar\omega_0 >> k_BT$ to ensure that the system is close to the ground state of the coupled superconductor problem. Such a condition is easily satisfied for small-grain metal systems, but may not be for more macroscopic junction structures.[3] If this condition is not satisfied, then the threshold criterion will depend on junction parameters such as L and C, and on temperature.[14]

Although the equations of motion of the RSJ model are quasiclassical, Eq. (1) implies that ϕ and n are really quantum-mechanical variables. In a full quantum theory we must also satisfy the following independent uncertainty relation:

$$\Delta E \Delta t \gtrsim \hbar. \qquad (2)$$

Both Eqs. (1) and (2) must be satisfied for the phase to be a well defined macroscopic quantum variable. In a parallel LRC circuit, the width of the resonance is $\gamma = 1/RC$. To measure a spontaneous classical fluctuation, which will occur within a frequency range γ, we require measurement over a characteristic time interval $\Delta t < \gamma^{-1}$. Over this time Δt, a phase uncertainty given by $\Delta\phi = \gamma\Delta t = \Delta t/RC$ will develop. We note that $\Delta\phi \lesssim 1$ for two reasons: (1) If it were not, then as noted above, ϕ would not be a meaningful quantum mechanical variable. (2) If $\Delta\phi > 1$, then $\gamma\Delta t > 1$ and we would not have enough time to measure ΔE before the fluctuation had decayed.[15]

We can compute the energy uncertainty if ϕ and n exist (i.e., if there is phase coherence) from[16] $\Delta E = (2e)^2(\Delta n)^2/C$ together with Eq. (1) and the relation $\Delta\phi = \Delta t/RC$:

$$\Delta E \gtrsim [(2e)^2/4C](RC)^2/(\Delta t)^2. \qquad (3)$$

Equations (2) and (3) are plotted in the $\Delta E - 1/\Delta t$ plane as curves b and a in Fig. 2 along with the vertical line c, $1/\Delta t = (RC)^{-1}$. The intersections of curves a

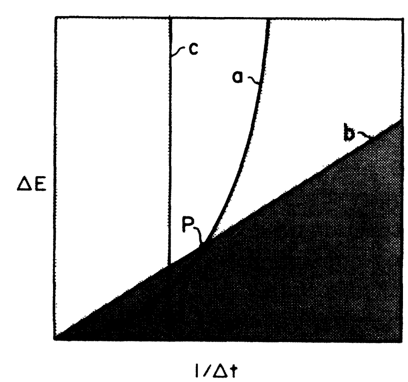

FIG. 2. Energy uncertainties ΔE vs $1/\Delta t$. Curve a is from Eq. (3) which depends on phase-number uncertainty and charging energy and curve b is from Eq. (2) which is the usual Heisenberg relation. The verticle line c corresponds to $\gamma\Delta t = \Delta\phi = 1$. The heavily shaded region is unphysical and the lightly shaded regions are not phase coherent.

and b are at the origin and at the point P where

$$(\Delta E)_P = \hbar^2\gamma/e^2R; \quad (\Delta t)_P = e^2R/\gamma\hbar. \tag{4}$$

This intersection P can occur either to the left or to the right of the line c. If $1/\Delta t \gtrsim \gamma$, i.e., if P is to the right of c, then all states compatible with Eq. (2) also satisfy Eq. (1). If P is to the left of c, i.e., $1/\Delta t < \gamma$, then there are states satisfying (2) which violate (1).

The Heisenberg uncertainty relation (2) means that the system is being described as a mixture of energy eigenstates, with energies $E > E_0$, the ground-state energy. When (1) is violated, the lowest-lying states are not the coherent ones, and the n-ϕ representation breaks down. In other words, the state of minimum internal energy is then one in which there is no Josephson coupling. Superconducting coupling is thus found only when

$$R \lesssim \hbar/e^2, \tag{5}$$

which is the condition for P being to the right of c. This can be interpreted as a threshold criterion for a tunneling resistance R_c above which zero-resistance Josephson coupling is not found.

If a thin granular film can be modeled as a square lattice of junctions, then the above condition in a single junction will also be the condition on the sheet resistance of the array. However, an actual thin-film geometry is more properly modeled as a *random* network of Josephson junctions. The sheet resistance of such a network has been described by Ambegaokar, Halperin, and Langer[17] and Levy *et al.*[18] using an argument which we briefly recapitulate. First disconnect all the junctions in the random network, and then reconnect them one by one in ascending order of resistance. A stage will be reached where the next junction completes an infinite cluster connecting the ends of the network. Let the normal-state resistance of this last junction be R_p. The measured normal-state sheet resistance of the entire two-dimensional network will be R_p, as this junction is the bottleneck. Junctions with resistances greater than R_p are irrelevant since they are always shunted by junctions with resistances of order R_p. Junctions with $R < R_p$ only form finite clusters; over macroscopic distances they do not affect the conductivity because the current must still pass through junctions with resistances of the order of R_p to get from one cluster to the next. If we identify R_c, the resistance of a single junction at the onset of superconducting coupling, with R_p, then Eq. (5) is the condition for global superconducting coupling. However, even if Eq. (5) is violated, this does not preclude local superconductivity.

The above resistance threshold $\hbar/e^2$ is then seen to be remarkably close to the observed threshold points of Fig. 1 which bracket $1.5\hbar/e^2$. The closeness of the agreement is remarkable since neither the uncertainty-principle argument nor the percolation argument purports to be precise. The main point of the above discussion is that the capacitance between clusters or grains drops out of the final result even though the concept of charge fluctuations is still central. The experimental data used here support our view that there is a universal threshold and that the value of the capacitance is irrelevant. Within the context of our model the latter will be true as long as $\hbar\omega_0 \gg k_BT$ and $E_1 \gg e^2/C$. If we take the limiting junction resistance to be $\hbar/e^2$, this implies that 1.6×10^{-16} F $\ll C \ll 3\times10^{-11}$ F. This is easily satisfied if C is the intercluster capacitance of the limiting junction of a film near its percolation threshold.

It is important to note that the above considerations also only apply when the Josephson coupling energy $E_1 > k_BT$; otherwise the coupling will be weakened by thermal fluctuations. This condition can be satisfied when $R = R_c$ not far below T_c for weak-coupling superconductors. In addition, effects due to collective plasma oscillations coupling many clusters are neglected. Presumably these would be of higher energy than the $k = 0$ mode which is implicit in our treatment of each junction as independent.

The above model is limited to the consideration of the coupling of sites where local superconducting order is already established. In the case of the Sn films in the present work the transition temperature of the clusters is very close to that of bulk material. This implies that the material within the clusters is highly ordered and localization in the conventional sense is not relevant. On the other hand, the absence of superconductivity in films of high sheet resistance has been attributed to pair breaking or to localization effects.[6–9] It is possible that either of these mechanisms could lower the local transition temperature and indeed the lowering of the transition temperature has been the focus of the interpretation of most of the previous data.[6–9] However, as long as local order survives, the above considerations should be valid provided the local transition temperature has not been driven to zero. Furthermore, in the context of the data reported here, it is difficult to understand how pair breaking or localization effects could disrupt *global* phase coherence without disrupting *local* superconductivity. The latter is present in the case of the Sn films we have studied, even when the low-temperature resistance is nonzero, i.e., when there is no global phase coherence.[13]

A somewhat surprising conclusion would follow from the existence of a universal resistance threshold if the quasiparticle resistance were the appropriate one in Eq. (5). First, for a single junction for which $\hbar\omega_0 \gg k_BT$, even if the quasiparticle resistance were low enough to permit coupling at some temperature, if it behaved in ideal manner, it would increase as T is reduced until the Josephson effect disappeared at a low enough temperature. A granular film could also reenter the normal state as a result of the increase in the intergrain quasiparticle tunneling resistances with decreasing T.

We speculate that a continuous film can be described in the continuum limit of an array of RSJ-coupled superconducting sites. A consequence of this generalization may be the universal disappearance of superconductivity in two dimensions as $T \rightarrow 0$, since for all metal films in two dimensions $R \rightarrow \infty$ as $T \rightarrow 0$.

In conclusion, we have observed that the onset of global phase coherence in two-dimensional granular superconductors seems to depend only on the sheet resistance. We have further presented a model in which the zero-point fluctuations of the junctions decouple the clusters of the film, thereby producing just such a result. The model is a generalization of the Anderson-Abeles idea[3,4] that the occurrence of superconductivity in granular films is intimately connected with charge fluctuations.

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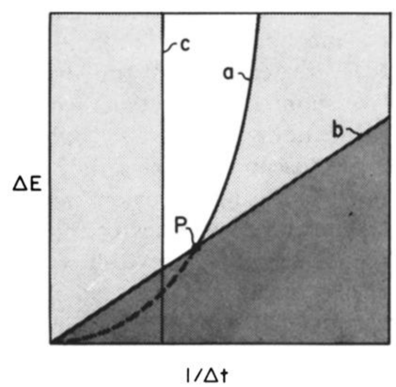

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