Quantum Phase Slips and Superconductivity in Granular Films

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(Received 21 February 1986)

An explanation is proposed for the recent observation that in ultrathin Sn films there is apparently a universal normal-state sheet resistance above which superconductivity cannot be established. It is demonstrated that when the sheet resistance is larger than the quantum of resistance, $R_Q = h/4e^2$, the order parameter suffers phase slips due to quantum tunneling driving the film normal at low temperature. This value of the resistance agrees quantitatively with experiment.

PACS numbers: 74.50.+r, 74.20.De, 74.40.+k

In a recent Letter Orr, Jaeger, Goldman, and Kuper[1] (OJGK) studied the onset of superconductivity in thin granular Sn films. They found the remarkable result that the only relevant variable in determining whether the film would become superconducting was the normal-state sheet resistance, R_N. Similar behavior had also been reported earlier.[2-5] For films with R_N slightly smaller than 6 k$\Omega/\square$ the film resistance was found to drop rapidly to zero near the bulk superconducting transition temperature for Sn, $T_c = 3.7$ K. In thinner films with R_N slightly larger than 6 k$\Omega/\square$, the resistance also dropped rapidly when cooled below T_c but upon further cooling reached a minimum and started *increasing*. This quasireentrant behavior had also been reported in several earlier experiments.[3,5] The most surprising aspect of the OJGK data was that in each of seven different samples, the *same* resistance was found to separate the two regimes. This led OJGK to suggest the existence of a universal maximum resistance for thin films, above which superconductivity cannot be established.

Superconducting granular films are often modeled as an array of superconducting islands linked together by Josephson junctions. Abeles[6] argued that charging effects in such junctions should be important and suggested that when the charging energy was larger than the Josephson coupling energy, quantum fluctuations should drive the junctions normal. This argument predicts a maximum sheet resistance which depends sensitively on the junction capacitance and would be expected to vary appreciably from sample to sample.

In this work I propose a possible explanation explanation for the apparently universal maximum resistance observed experimentally. By combining the picture of an array of islands joined by Josephson-junction links with recent calculations relevant to the dissipative quantum dynamics of a single junction,[7-9] a natural explanation for the observed data is presented. In contrast to previous work on granular films[10-12] the Josephson-junction links are assumed to be shunted by a normal resistance. This dissipative element plays a crucial role in modifying the junction dynamics. For junctions with shunting resistances, R_s, larger than the quantum of resistance $R_Q = h/4e^2 = 6.45$ kΩ, quantum tunneling induces phase slips which drive the junctions normal. In contrast, for $R_s < R_Q$ the dissipation is sufficient to stabilize this quantum phase slip and the junctions become superconducting at low temperatures. This behavior manifests itself in the sheet resistance. The approach described in this work not only gives the correct value for the observed maximum resistance but also provides a natural explanation for the quasireentrant behavior.[13]

The Sn film is visualized as a random network of islands, which have a normal-state resistance substantially smaller than that of the film. These islands are connected to neighboring islands by high-resistance links which depend on the local detailed geometry and are presumably rather widely distributed. The large change[5] in R_N with additional deposition of Sn is accounted for naturally by the expected sensitivity of a link resistance to such deposition. The islands are assumed to be larger than the BCS coherence length and sufficiently ordered that they become superconducting near the bulk T_c of Sn. The adjoining links can then be thought of as small Josephson junctions.

Within this model the sheet resistance can be estimated from knowledge of the resistance of the individual links by use of an argument due to Ambegaokar, Halperin, and Langer.[14,15] Imagine disconnecting the resistors in descending order starting with the largest resistors first. At some resistance R_p, the network of interconnected islands reaches the percolation point; removal of the next resistor destroys conduction since the connected cluster of islands no longer reaches across the sample. The sheet resistance should then be given approximately by R_p since resistors of this value are bottlenecks for conduction and the larger resistors tend to be shorted out by the infinite cluster. The above argument only applies in two dimensions (2D); in 3D, for example, the resistance (and resistivity) of a network will depend sensitively on the average spacing between islands, even for a given realization of resistors.

From the above considerations it is apparent that an understanding of the behavior of the film's sheet resis-

tance requires first an examination of the temperature dependence of the resistance of a *single* link. Consider then the behavior of a single small Josephson junction. I assume that the classical dynamics of the junction is described by the so-called resistively shunted junction model.[16] For a junction biased with current I the phase difference ϕ of the superconducting order parameter across the junction satisfies

$$C\ddot{\phi}+\dot{\phi}/R_s+(2e/\hbar)^2U'(\phi)=(2e/\hbar)\xi(t), \quad (1)$$

$$U(\phi)=-E_J\cos\phi-(\hbar/2e)I\phi, \quad (2)$$

with C the junction capacitance, R_s a normal resistor shunting the junction, E_J the Josephson coupling energy, and $\xi(t)$ Gaussian noise with mean zero and variance $2k_BT/R_s$. I assume that R_s is *independent* of temperature.[17,18] Of interest is the actual junction resistance $R(T)$, defined in terms of the voltage V across the junction as $R(T)=dV/dI$ at $I=0$. This resistance should not be confused with R_s.

Equation (1) is equivalent to the classical dynamics of a fictitious particle with damping R_s^{-1} moving in the potential $U(\phi)$. Since $V=\hbar\dot{\phi}/2e$, the junction resistance is proportional to the *mobility* of the particle down the potential. Above T_c, E_J vanishes and $R(T)$ is equal to R_s. Upon cooling, two effects conspire to reduce $R(T)$. The coupling energy increases rapidly near T_c providing higher barriers in the potential. Moreover, the fluctuating force becomes less effective at surmounting these barriers. If quantum effects in ϕ were ignored, the junction resistance would plummet exponentially fast to zero and vanish completely at $T=0$. Inclusion of quantum fluctuations could drastically modify this behavior. At low temperatures one might expect that ϕ could tunnel quantum mechanically between adjacent minima in the potential, enhancing the mobility and perhaps giving a nonzero junction resistance at $T=0$. In this scenario, a finite voltage would develop across the junction as a result of successive phase slips (of 2π) induced by quantum tunneling.

To quantify the role played by quantum fluctuations a fully quantum treatment of the junction dynamics is necessary. I adopt the theoretical framework pioneered by Caldeira and Leggett[17] who considered a Lagrangean of the form

$$L_{CL}=\tfrac{1}{2}C(\hbar\dot{\phi}/2e)^2-U(\phi)+\textstyle\sum_j\frac{1}{2}m_j\dot{x}_j^2-\sum_j\frac{1}{2}m_j\omega_j^2(x_j+\lambda_j\hbar\phi/2em_j\omega_j)^2, \quad (3)$$

where ϕ and the oscillator coordinates x_j are quantum degrees of freedom. The dissipation due to the shunting resistor is modeled by the bath of harmonic oscillators. The coupling constants λ_j are chosen[17] so that in the classical limit the dynamics described by L_{CL} reduces to the resistively shunted junction model, Eq. (1),

$$\textstyle\sum_j(\pi\lambda_j^2/2m_j)\delta(\omega-\omega_j)=R_s^{-1}. \quad (4)$$

Since ϕ is a phase variable, it is only defined on the interval 0 to 2π. However, the Lagrangean (3) is not invariant under $\phi\rightarrow\phi+2\pi$. For processes where ϕ changes by a large amount with respect to 2π, such as the voltage response to a bias current which involves ϕ cycling through many rotations, a more appropriate Lagrangean is[19,20]

$$L=\tfrac{1}{2}C\left(\frac{\hbar\dot{\phi}}{2e}\right)^2+E_J\cos\phi-\frac{\hbar\dot{\phi}}{2e}[\textstyle\sum_j\lambda_jx_j+It]+L_{osc}, \quad (5)$$

where L_{osc} is the Lagrangean for the uncoupled oscillator bath. The classical dynamics of ϕ generated by (3) and (5) are entirely equivalent. However, (5) is invariant under $\phi\rightarrow\phi+2\pi$. By analyzing the formal quantum expressions for the voltage $\dot{\phi}$ in response to the driving current I, one can show[20] that so long as R_s^{-1} is nonzero the result obtained from (5), for ϕ restricted to the interval 0 to 2π, is equivalent to that obtained from (3) for ϕ unrestricted ($-\infty<\phi<+\infty$). Thus for purposes of calculating the junction resistance, L_{CL} can be safely applied.

For an unbiased washboard potential, the Caldeira-Leggett model with $\phi\in[-\infty,\infty]$ has a $T=0$ localization transition first pointed out by Schmid,[7] which has since been studied in detail.[8,9] The transition can be analyzed by expressing the partition function for the full system (2) as a path integral in imaginary time over paths $\phi(\tau)$ and $x_j(\tau)$. After integrating out the harmonic oscillator degrees of freedom and taking β ($=1/kT$) to infinity one finds[8] $Z\sim\int D\phi(\tau)\times\exp(-S)$ with

$$S=(\alpha/8\pi^2)\int d\omega(|\omega|+\omega^2/\lambda)|\phi(\omega)|^2-E_0\Lambda\int d\tau\cos\phi(\tau), \quad (6)$$

where $\Lambda=(R_sC)^{-1}$ and $E_0=E_J/h\Lambda$ is a dimensionless coupling energy. Here α^{-1} is the dimensionless shunting resistance measured in units of R_Q,

$$\alpha=R_Q/R_s, \quad R_q=h/4e^2, \quad (7)$$

and $\phi(\omega)$ is the Fourier transform of $\phi(\tau)$. In (6) Λ plays the role of a high-frequency cutoff. A perturbative renormalization group, in powers of E_0, can be implemented to study the phase transition. The resulting differential flow equations are[8] ($dl=-d\Lambda/\Lambda$)

$$dE_0/dl=(1-\alpha^{-1})E_0(l)+O(E_0^3), \quad (8)$$

and $d\alpha/dl=0$, to all orders in E_0. Flows can also be

obtained in the tight-binding ($E_0 \to \infty$) limit. One finds that when $\alpha < 1$, E_0 is irrelevant and scales to zero at long "time" scales indicating that the ground-state wave function is extended in the coordinate ϕ. For $\alpha > 1$, on the other hand, the periodic potential is relevant. The discrete translational symmetry of the potential is broken and the wave function is localized. In this case the dissipation due to R_s is sufficiently strong that all quantum tunneling between minima in the potential is completely suppressed. Crucial to the present discussion is that the location of the transition is determined only by the shunting resistance, R_s. The coupling energy and the junction capacitance play no role.[21] Moreover the value of R_s at the transition is precisely $R_Q = h/4e^2$.

One can also study directly the dynamics[22] generated by L_{CL}. Recently, in collaboration with Zwerger,[8] I have calculated the particle's steady-state mobility in *real time*. From this one can infer the junction resistance, $R(T)$. To leading order in the coupling energy, E_J, $R(T)$ is given by

$$R(T)/R_s = 1 - (\pi/\alpha\hbar k_B T)E_J^2 \int_0^\infty dt\, \mathrm{Re}\exp[P(t) - iQ(t)], \tag{9a}$$

$$P(t) = (2/\alpha)\int_0^\infty d\omega\, \omega^{-1}(\cos\omega t - 1)\coth(\tfrac{1}{2}\beta\hbar\omega) f(\omega/\Lambda), \tag{9b}$$

with $Q(t)$ the $\beta \to 0$ limit of $\frac{1}{2}\hbar\beta\, \partial_t P(t)$ and $f(x) = (1+x^2)^{-1}$. The temperature dependence enters into $P(t)$ and also via E_J, which depends implicitly on T. When $R_s > R_Q$, Eq. (9) is useful for arbitrary temperature,[8] but it breaks down as $T \to 0$ when $R_s < R_Q$.

By analysis of (9) and in light of the $T = 0$ transition the following picture emerges. In the normal state, the junction resistance is equal to the shunting resistance. Upon cooling below T_c, $R(T)$ falls since E_J grows rapidly. Moreover, the thermal bath becomes less effective at kicking the particle over the washboard barriers. At low temperatures the behavior depends critically on R_s. When $R_s < R_Q$, the junction resistance vanishes at $T = 0$, since the ground state is localized and the particle immobile. In this case $R(T)$ is a monotonic function of the temperature which is negligibly small for T well below T_c. On the other hand, when $R_s > R_Q$ and the ground state is extended, an evaluation of the integrals in (9) reveals a nonmonotonic temperature dependence of $R(T)$. Below a crossover temperature T^*, $R(T)$ *increases* with decreasing temperature and as $T \to 0$ it approaches a nonzero constant, $R(T=0) = R_s$. This crossover behavior is due to *quantum tunneling* of ϕ. At high temperatures the motion is classical in nature involving over-the-barrier transport. Below T^*, however, quantum tunneling starts to dominate. In this regime, decreasing the temperature leads to a *larger* resistance since the environment become less effective at destroying the quantum coherence necessary for tunneling. The crossover temperature will in general depend on the junction capacitance, the coupling energy, and the shunting resistance. In the limit that T^* is considerably less than T_c, so that E_J is weakly temperature dependent, and in the limit of small E_J, T^* is given in terms of the normal-state properties by[8] $T^* = g(\alpha)e^2/3C$, where $g(\alpha)$ is a monotonic function which approaches 0 as $\alpha \to 1^-$ and 1 as $\alpha \to 0$. With a reasonable value of the capacitance, $C \sim 10^{-15}$ F, this is in the range of the observed crossover temperature.

The central results obtained above can be summarized as follows. For junctions with shunting resistances larger than R_Q, the phase will slip at low temperatures as result of quantum tunneling. The junction resistance will exhibit a quasireentrant behavior, rising at low T, and will approach a nonzero constant as $T \to 0$. In contrast, when $R_s < R_Q$, the dissipation is sufficiently strong to entirely suppress quantum tunneling of the phase. Only thermally activated phase slip will be present and for T well below T_c the junction will have negligible resistance.

Consider now the behavior of the film resistance when each link in the network of islands is modeled as a shunted Josephson junction. As above I assume R_s to be temperature independent.[23] Divide all the links into two groups, a high- and a low-resistance group with R_s larger and smaller, respectively, than the quantum of resistance R_Q. There are two possible scenarios:

(i) The network of islands interconnected by the low-resistance group alone forms a percolating cluster which extends across the entire film. As the film is cooled below T_c, the low-resistance links become superconducting. The cluster of islands interconnected by these links then forms a superconducting network percolating across the film.[24] All the other resistors will be shorted out by this cluster and the film as a whole should carry current with no resistive losses.

(ii) The network of islands joined by the low resistors does not percolate, but forms finite clusters connected to one another by high resistors. As the film is cooled below T_c, these finite clusters turn superconducting. However, the film resistance will be dominated by the high resistors which form bottlenecks between the clusters. As we have seen, the resistance of these links will also initially decrease upon cooling below T_c. At sufficiently low temperature, however, quantum tunneling will induce phase slips across the links, leading to an increasing resistance. The film

resistance should then be a nonmonotonic function of temperature exhibiting a quasireentrant behavior and a resistive state at the lowest temperatures.

The above picture should also hold for granular systems of higher dimensionality. However, it is only in 2D that the *normal*-state (sheet) resistance R_N, in the two cases above, will be insensitive to the average spacing between islands. Specifically, for a 2D film, the Ambegaokar-Halperin-Langer percolation argument[14,15] indicates that in case (i) R_N will typically be smaller than R_Q, whereas in case (ii) it will be larger. Thus films with $R_N < R_Q$ should exhibit superconductivity at low temperatures whereas films with $R_N > R_Q$ should show a quasireentrant behavior with a finite low-temperature resistance.

I believe that the above arguments provide an extremely natural explanation for the experiments by OJGK.[1] Consistent with the experiments, R_N emerges as the only relevant variable in determining the film's low-temperature behavior. Moreover the resistance which separates the two low-temperature regions, $R_Q = h/4e^2$, agrees quantitatively with the measured value. In addition, for $R_N > R_Q$ the theoretical calculations reproduce the observed quasireentrant behavior.

It is a pleasure to acknowledge stimulating discussions with S. Chakravarty, D. S. Fisher, M. E. Fisher, Y. Fu, A. M. Goldman, A. J. Leggett, O. Martin, and B. G. Orr. I am particularly indebted to B. G. Orr for bringing to my attention the experiments on Sn films. I am grateful to AT&T Bell Laboratories for financial support. This work was also supported in part by the John D. and Catherine T. MacArthur Foundation at the University of Illinois under Grant No. 0-6-40129.

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[23]This assumption is crucial since it enables one to relate the low-T behavior of a junction to its normal-state resistance.

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