RAPID COMMUNICATIONS

PHYSICAL REVIEW B VOLUME 32, NUMBER 11 1 DECEMBER 1985

Local superconductivity in ultrathin Sn films

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(Received 30 September 1985)

The thickness dependence of superconductivity has been investigated in ultrathin Sn films near the threshold for normal-state conduction. An evolution from insulating to metallic superconducting behavior is observed with increasing thickness and decreasing sheet resistance. A striking feature of the data is the observation of quasireentrant superconductivity which may be a consequence of the existence of local rather than global superconducting order.

The conditions for the onset of bulk solid behavior and the development of associated cooperative phenomena such as ferromagnetism and superconductivity in ultrathin films are important problems in condensed matter physics. In the case of superconductivity, the decrease of the local transition temperature with decreasing thickness and increasing sheet resistance has been studied in amorphous alloy films with fixed disorder[1] and modeled using the theory of localization.[2] The actual phase transition of two-dimensional superconducting systems has been found to be a topological transition involving the unbinding of superconducting vortices of opposite helicity.[3,4]

A problem different from the development of superconductivity in essentially homogeneous systems is its onset in films in which there is a change in the morphology from a structure consisting of isolated clusters to one consisting of connected clusters. This connectedness transition and its relationship to the normal-state conductivity has been successfully modeled using percolation theory.[5,6]

In this article we report on the nature of the superconducting state in Sn films which exhibit a connectedness transition. We have documented the systematic evolution from insulating to metallic behavior as the sheet resistance of a film is decreased by increasing its thickness. We can identify characteristically different regions in this evolution which takes place over a very narrow range of only a few angstrom change in film thickness. The most striking feature of the data is the appearance of a quasireentrant state in films slightly thinner than those in which full superconductivity (zero or near zero resistance) appears. Quasireentrant behavior, which we will call local superconductivity, in contrast with the global superconducting behavior of the zero-resistance state, has been reported in some films of granular Sn,[7] Al,[8] and Hg-Xe.[9] In the present instance the observation of the systematic dependence of the phenomenon on sheet resistance and thickness presented here should greatly aid in the development of a detailed model.

Sn films were evaporated onto a glazed alumina substrate held at 18 K through a mask which defined the film dimensions as 5.0×0.5 mm^2.[10] The vapor source was a molecular-beam oven. The vapor flux density was estimated to be uniform to better than one part in 4000 over the area of the sample. Measurable conductance was obtained after the deposition of between 15 and 30 Å of material. Deposition was then stopped and the sample moved while in the ultrahigh-vacuum environment ($\simeq 5\times 10^{-10}$ Torr or lower) into a thermally shielded environment for more detailed study. The sample could then be returned to the growth chamber for the deposition of additional material. In this manner the sample thickness and associated properties could be changed in a semicontinuous manner. Sample thicknesses are nominal and are computed from the product of the deposition time and rate. The latter was determined from a long exposure to the vapor source of a calibrated crystal-oscillator monitor. Resistance was measured using a nanovoltmeter and a picoampere current source. Typically one nanoampere was used in the measurement of the high-resistance samples. As long as the sample temperature remained below the highest temperature during deposition, the measured resistance was not subject to annealing effects.

In Fig. 1 the resistance versus temperature is plotted for two sets of films produced by incrementing the thickness in the manner described above. It should be noted that the normal-state resistance changes by a factor of 200 while the total thickness deposited increases by less than a nominal 1.5 Å. At high temperatures the resistance is seen to rise more rapidly than logarithmically in the normal state for films which do not become superconducting at low temperatures. This gives way to a logarithmic increase in the resistance with temperature when the normal resistance falls to the order of $\hbar/e^2$ $\Omega/\square$. Also evident in the high-resistance samples is a point of very rapid increase in the rate of rise of the resistance as the temperature is decreased through the bulk superconducting transition temperature of Sn. This "kink" evolves into a minimum in the resistance as the sample is made thicker.

With increasing thickness the films enter a regime where they seem to undergo a true superconducting transition, but on close inspection actually exhibit a nonzero resistance down to the lowest temperatures measured. (This behavior is not shown in Fig. 1.) Finally, in even thicker films and only when the sheet resistance falls to the order of $1.5\ \hbar/e^2$ the samples show true superconductivity, or global superconductivity, i.e., zero resistance at temperatures near the bulk T_c of Sn. This behavior was found in seven sets of films and was independent of the thickness at which the films were continuous.

We have found that a rather elementary percolation simulation of superconducting clusters similar to one first discussed by Simanek[11] can reproduce the qualitative features of the reentrant behavior discussed above. What is involved in the present instance is percolation of the Josephson coupling between superconducting sites on a random network. In our idealized simulations we vary the strength of the coupling and the percentage coverage of Josephson-like bonds. Above T_c the bond lattice is randomly filled with normal-state resistances. A renormalization procedure is used to find the overall conductance of the sample. Below T_c this lattice becomes a random distribution of tunneling junctions. The temperature dependence of the lattice conductance is then given by the net effect of the temperature dependences of individual bonds. For the Josephson-like channels this is given by the thermal rounding of the resistance in the limit of zero bias.[12] The dynamical resistance in this model of fluctuation rounding of the I-V characteristic is given by the resistance of the quasiparticle tunneling channel which is in parallel with the Josephson channel.

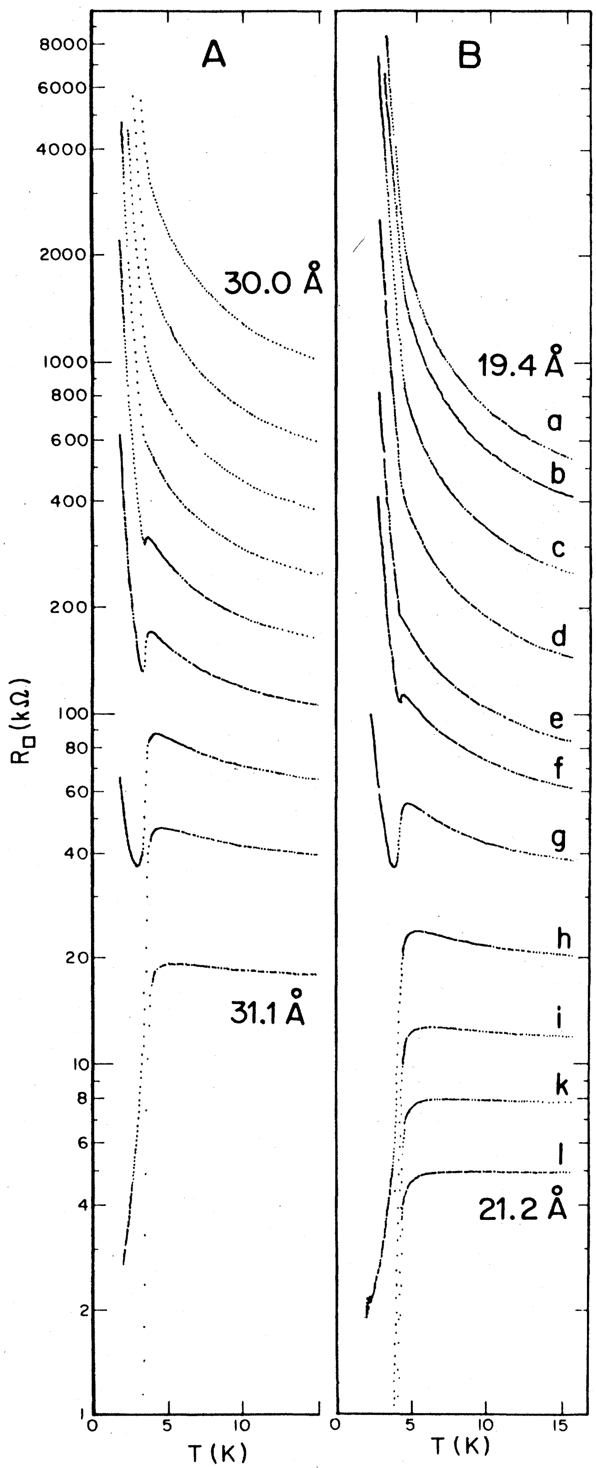

FIG. 1. Resistance vs temperature $R(T)$ for two sets of Sn films. Typical thickness increments are 0.1 Å. The small letters correspond to Figs. 2 and 3.

Because of the idealized character of the model we will discuss its predictions qualitatively rather than attempt to fit to the data. As the temperature is lowered below the local T_c of the clusters all quasiparticle tunneling channels gradually freeze out and the quasiparticle current becomes very small. Although the simulation was carried out taking the temperature dependence of the energy gap to be that of pure Sn, and omitting amplitude fluctuations of the order parameter, all of the features of the temperature dependence of the sets of films in Fig. 1 are qualitatively reproduced. When the coverage of the Josephson bonds exceeds the percolation threshold in two dimensions ($p_c = 0.5$), the temperature coefficient of resistance (TCR) at low temperatures changes sign. For lower coverages the simulation reproduces the "kink" which develops into a minimum, with increasing coverage of Josephson bonds, at a temperature close to the bulk transition temperature. It should be noted that for weak-coupling systems there would be no dramatic change in behavior at p_c, and at higher coverages the transition temperature would be greatly depressed. This type of behavior has been seen in many types of oxidized metallic granular films, and appears to be a reason that quasireentrant behavior is not observed in those cases.

The applicability of this percolation picture of local superconductivity is qualitatively supported by the low-temperature I-V characteristics of the films which are shown in Fig. 2. As the sample thickness is increased an abrupt change in curvature occurs. High-resistance samples have I-V characteristics with curvatures like those of junctions with electrodes characterized by a smeared energy gap. Lower resistance samples have I-V characteristics with cur-

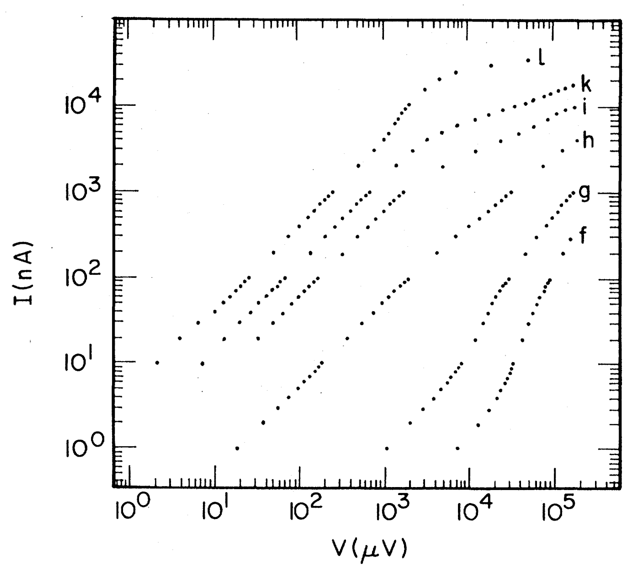

FIG. 2. Current vs voltage characteristics, $I(V)$, at 2.5 K. The change in curvature coincides with the change in the low-temperature behavior of films given in Fig. 1.

vatures like those of thermally rounded Josephson junctions which have a hint of a critical current. This change in behavior occurs at precisely the thickness at which the temperature coefficient of resistance (TCR) at low temperatures changes sign. Within a percolation model these are consistent behaviors. In each instance the TCR and the I-V characteristics are appropriate for the type of bond which spans the sample. Also in detailed agreement with the model is the temperature dependence of the resistance in the regime in which quasiparticle tunneling dominates. Figure 3 shows a set of $R(T)$ curves for high-resistance films normalized at 3 K along with a theoretical curve for $R(T)$ in the limit of zero bias computed from the single-particle tunneling conductance of a superconductor-insulator-superconductor junction. At low temperatures, where the effects of fluctuations are relatively unimportant, the agreement is extremely good.

The above percolation model of tunneling junctions on a lattice is, as has been mentioned, a major simplification of the geometry of these materials. The films are actually a random material consisting of metallic clusters connected by many random tunneling links in parallel with various metallic links. As a film becomes thicker global superconductivity is established along with metallic connectedness in the normal state. When the latter happens, tunneling, which appears to dominate the percolation model of the onset of superconductivity, becomes irrelevant. A more conventional percolation picture is then a good description of the normal-state resistance. Indeed, we find that near, but beyond, the percolation threshold for normal conductivity the latter scales as $(p-p_c)^{\mu}$ with an exponent $\mu = 1.36 \pm 0.06$, if we assume the areal coverage p to be proportional to the nominal thickness.[13] We find this relationship to be satisfied in the normal state of our films over a range of thicknesses from that at which a conductivity threshold is found to about twice that value. These observations support the relevance of a percolation picture to the normal

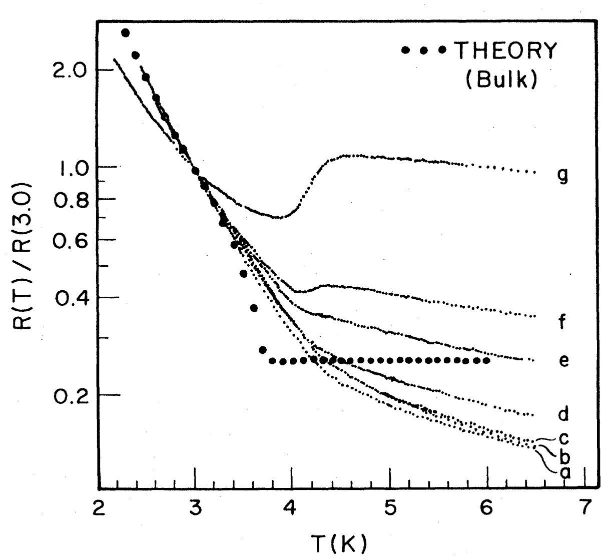

FIG. 3. Low-temperature resistance vs temperature normalized to 3 K. Films a–f asymptotically show identical low-temperature behavior which is in agreement with a model which attributes the low-temperature resistance to the freezing out of quasiparticle excitations (dotted line).

conduction. One must be careful to note that the percolation of the superconductivity and the percolation of the normal conduction are separate problems and may, as in this instance, have different critical thresholds.

It is important to point out that global superconductivity is observed when the normal-state resistance increases logarithmically with decreasing temperature but is not found when the increase is faster than logarithmic. This implies that superconductivity is compatible with weak but not strong localization.

It should be noted that other explanations have been suggested for the resistance minima which we have described within a percolation model. They may be a consequence of zero-point charge fluctuations between Josephson-coupled clusters as originally suggested by Abeles.[14] The correlated mean-field treatment of this problem recently given by Fazekas, Muhlschlegel, and Schroter[15] which neglects vortex excitations, attributes the resistance minima to maxima found in a correlation function which is a measure of the intergrain phase coherence. However, this model makes no quantitative predictions for $R(T)$. Another possibility is that quasireentrance is due to a low-temperature vortex-unbinding transition driven by zero-point charge fluctuations.[16] This appears to be ruled out by the observation that the resistance minima occur at values of the sheet resistance which are too high to be produced by flux-flow resistance alone as would be required in this model. Nevertheless, such a picture may describe films with normal-state resistances slightly above the threshold for zero resistance or global superconductivity, where a relatively small nonzero resistance is exhibited down to the lowest temperatures studied. Further work involving the extension of the measurements to lower temperatures may clarify the nature of this behavior.

The behavior which we have described here has been characterized as local superconductivity because the superconducting properties do not span the sample. The superconducting sites, which are the metallic clusters of the coalescing film are coupled by superconducting tunneling junction bonds. The progression towards global superconductivity is associated with a spread across the film of connectedness with zero resistance which can be qualitatively modeled using concepts from percolation theory.

It is clear that a truly quantitative description of the continuous evolution of superconductivity with increasing thickness in a nucleating film must treat the effects of randomness in a more microscopic fashion than we have and in addition include the possibility of an XY-like phase transition of the clusters involving zero-point charge fluctuations. Localization is clearly relevant near the threshold for global superconductivity given the correlation of the observation of superconductivity with the nature of the localized behavior of the normal state.

The authors would like to thank Dr. J. Gordon and Dr. J. Maps for helpful discussions. They would also like to acknowledge useful discussions with J. Jose, S. Doniach, M. Ma, P. Fazekas, R. Orbach, G. Deutscher, Y. Imry, J. Straley, and G. Stanley. B.G.O. would like to thank the IBM Corporation for financial support. This work was supported in part by the National Science Foundation Low Temperature Physics Program under Grant No. NSF/DMR 8503087, and by the Microelectronics and Information Sciences Center of the University of Minnesota.

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