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Threshold for superconductivity in ultrathin amorphous gallium films

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Systematic studies of the onset of superconductivity in ultrathin amorphous Ga films have revealed the existence of a threshold dependent only on the normal-state sheet resistance. Global superconductivity, or zero resistance, develops only when the normal-state sheet resistance falls below 6000 Ω/□. This result agrees with previous observations on crystalline Sn films and further supports the notion of a universal resistance threshold.

In a recent Letter[1] we reported that the condition for the onset of superconductivity in ultrathin Sn films appeared to depend only on their sheet resistances in the normal state, with a threshold occurring when they fell below about 6000 Ω/□. This conclusion resulted from investigations[2] in which the evolution of the electrical properties of films were studied using a technique in which small amounts of additional material could be added to a surface effectively building up a continuous film from isolated metallic clusters on the substrate. The apparatus[3] used to do this permitted studies of electrical properties to be carried out between successive depositions. Using increments of thickness which were at most 0.2 Å, it was observed that the last film exhibiting finite resistance at low temperature had a normal-state resistance of the order of 7 to 8 kΩ/□ whereas the first superconducting film had a normal-state resistance of 4 to 5 kΩ/□. The pairs of normal-state resistances for seven sets of films appeared to bracket 6000 kΩ/□. This apparent threshold for superconductivity was found at different nominal thicknesses, ranging from about 10 Å to more than 100 Å. This implies that the details of the film geometry, and in particular intercluster capacitance were not relevant variables in determining the onset of global superconductivity. The existence of superconductivity at low temperature seemed to depend only on the sheet resistance in the normal state.

The above remarkable result has been explained at least qualitatively in a number of related ways. An ultrathin film can be modeled as a network of metallic clusters coupled by Josephson tunneling junctions, and the existence of a resistance threshold for superconductivity in a junction will translate into a threshold for the entire network using an argument espoused by Ambegaokar, Halperin, and Langer.[4] The use of this argument does not mean we regard the film as actually reducing to a single junction. Junction threshold conditions which depend principally on their normal tunneling resistance can be derived in the overdamped limit of the resistively shunted junction (RSJ) model.[1,5-8] The relevant physical processes involve quantum fluctuations of the order-parameter phase difference in the presence of dissipation. The condition for superconductivity requires the localization of the phase difference across the junction, in one of the minima of the washboard potential of the RSJ model. Such a localization will occur when the normal-state resistance falls below a value of order $\hbar/e^2$. In an analysis which goes beyond a single-junction picture, Chakravarty, Ingold, Kivelson, and Luther[9] have shown that the threshold can be understood in terms of a dissipation-driven phase transition involving an array of junctions. Central to all of these explanations is the coupling of the macroscopic quantum-mechanical degree of freedom of the junction to dissipation.[10]

In this Communication we report an extension of our previous experimental work on Sn films[1] to films of Ga. With refinements in the experimental technique, our Ga data further support the notion that a sheet resistance very close to $\pi/2(\hbar/e^2)$ in the normal state is indeed a threshold normal resistance above which global superconductivity is not found. As in our previous discussions of this subject, we make an important distinction between global superconductivity, where the sample resistance is measured to be zero within experimental accuracy, and local superconductivity where there is evidence of change in the behavior of the system related to superconductivity, e.g., the occurrence of a minimum in the resistance versus temperature, $R(T)$. Because the Ga films of the present work are amorphous,[11] the results of the measurements may be used to clarify the role of localization and interaction effects in setting the threshold for the appearance of zero resistance or global superconductivity.

Gallium films were formed by evaporation onto glazed alumina substrates held at a temperature of less than 18 K in a vacuum of less than 3×10^{-10} Torr. The technique of studying the thickness dependence of low-temperature properties was refined to a level at which it was possible to deposit material in amounts corresponding to nominal thickness increments of 0.05 Å. As in previous experiments electrical measurements as a function of temperature and magnetic field were carried out between successive depositions of material. In contrast to the studies which were carried out on Sn films where the temperature did not go below 2.0 K, for the work on Ga the lowest temperature was 0.7 K.

Figure 1 shows the evolution of $R(T)$ of a Ga film with increasing nominal thickness. It contains the first 24 of more than 50 traces spanning five decades of normal-state

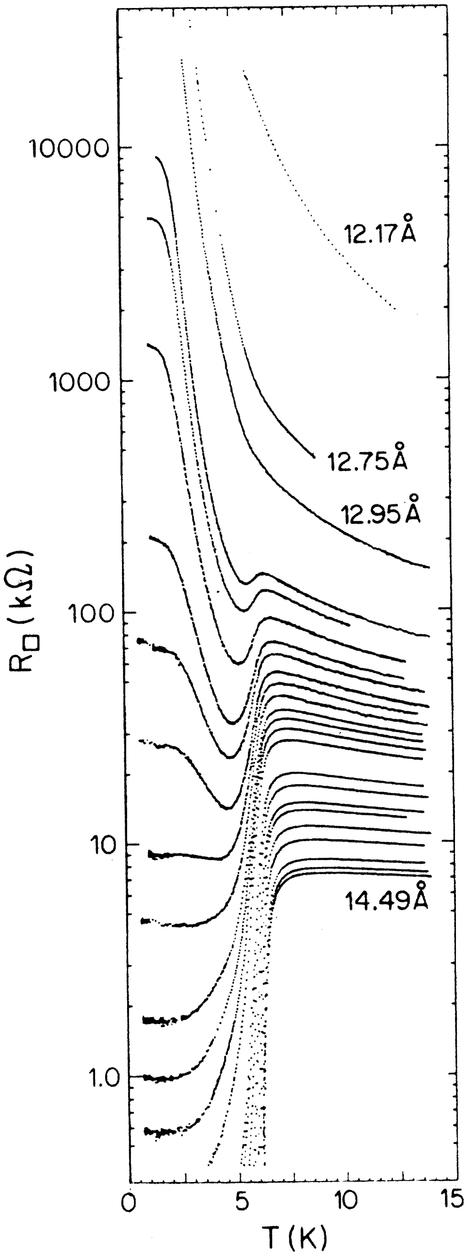

FIG. 1. Evolution of the resistive transition $R_\square(T)$ for a Ga film. The thickness increments between traces are typically 0.05–0.1 Å. Shown are the first 24 of more than 50 traces spanning five decades of normal-state resistances.

sheet resistance. With the exception of the top three traces which span a range of thickness from 12.2 to 12.95 Å, the increments of thickness between traces are between 0.05 and 0.1 Å. Measurements were carried out inside a magnetic shield which reduced the ambient field to a maximum of 0.01 G. The features of the transition in Ga greatly resemble those of Sn films reported earlier[1,2] with a number of important differences. In the previous work on high-resistance Sn films, $R(T)$ increased sharply with decreasing temperature apparently without limit, whereas in the present studies on Ga films $R(T)$ appears to flatten out at the lowest temperatures even when there is a minimum. These results suggest that the picture of quasireentrant behavior being caused by the freezing out of quasiparticle tunneling channels at the lowest temperatures[2] may have to be modified in some manner. Figure 1 shows that this minimum eventually gives way to a flat, temperature-independent tail as more material is added.

The data, in particular the observation of finite resistance tails, suggest another method of characterizing the onset of superconductivity as the thickness of a film is increased and its normal sheet resistance decreases. In Fig. 2, $\log R_\square$ (0.7 K) is plotted as a function of $\log R_\square$ (14.0 K), the latter being "defined" as the normal-state resistance. The resultant curve is a straight line until the 14 K value of the resistance is the order of 10 kΩ/□ at which point the 0.7 K value of the resistance drops sharply with further decrease in the 14 K resistance. The asymptotic value of the high-temperature resistance as the low-temperature resistance falls by many orders of magnitude is very close to $h/4e^2$ or $\pi/2(\hbar/e^2)$.

These measurements in essence bridge the gap between the resistances in the normal state corresponding to a finite low-temperature resistance, and those corresponding to zero resistance which were shown in Ref. 1. An onset resistance of the value displayed in Fig. 2 has been found in two different series of Ga films. In the other film the onset of superconductivity occurred at a nominal thickness of roughly 120 Å, in contrast to the 14.5 Å value for the data of Fig. 2. This difference implies rather different geometries for the two films at the critical resistance for superconductivity.

The fact that the onset of superconductivity is found at the same normal sheet resistance for rather different materials, amorphous Ga and crystalline Sn, over a substantial range of thicknesses, strongly suggests that there is a universal resistance threshold independent of both chemical and also geometrical parameters such as the sheet capacitance. As was mentioned above, the various models which exhibit a resistance-dependent threshold imply that the (inter-) cluster capacitance must be small. Although we have not measured the capacitance directly, the fact that the resistance threshold occurs for different effective

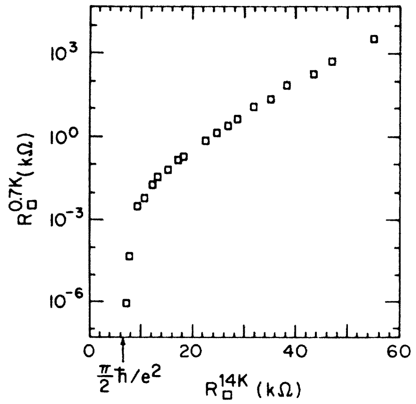

FIG. 2. Low-temperature resistance $R_\square(0.7\text{ K})$ vs normal-state resistance $R_\square(14.0\text{ K})$. $R_\square(0.7\text{ K})$ has dropped several orders of magnitude when global superconductivity sets in at $\sim\pi/2(\hbar/e^2)$.

thicknesses and thus different aspect ratios of the clusters which constitute the films strongly suggests that capacitance is not a relevant variable.

In our previously reported work on Sn films[1,2] it appeared as if localization and related interaction effects were essentially unrelated to the threshold condition for superconductivity. Furthermore, there was no significant reduction of T_c with increasing sheet resistance. In addition, there was no significant magnetoresistance at low fields. These results would appear to be in conflict with previous experimental and theoretical studies of the interplay between disorder and superconductivity in thin films.[12-14] The absence of magnetoresistance could be consistent either with an extremely short inelastic scattering length, or with no localization effects at all. The fact that T_c for Sn was not reduced substantially even in high sheet resistance films supports the latter. This is in contrast with the work of White, Dynes, and Garno[14] in which substantial magnetoresistance was observed in Sn films which were also deposited onto low-temperature substrates. The difference between the behavior of those films and the ones of our previous investigations must be the length scale of disorder, our Sn films being disordered on a larger length scale than those of White *et al.*

The studies of Ga films appear to provide an example of a situation in which the metal is disordered on a relatively short length scale. The reason for this is that Ga films deposited onto substrates held at low temperatures are known to be amorphous.[11] Indeed the identification of an amorphous phase is associated with a specific value of the superconducting transition temperature of 8.4 K. The various crystalline phases of Ga become superconducting at significantly lower temperatures.

The systematic variation of the transition temperature with sheet resistance in Ga films is of considerable interest in its own right. In Fig. 3 we plot the local transition temperature T_{cL}, arbitrarily defined to be the 50% point of the resistive transition, as a function of the logarithm of sheet resistance at 14 K. It is seen that this smooth curve extrapolates to a value in excess of 8 K, strongly suggesting that the films are actually amorphous with T_{cL} depressed from its bulk value by some microscopic process associated with the sheet resistance of the film. The observed logarithmic depression of T_{cL} is a weaker function of $R_\square$ than predicted by either electronic localization or the proximity effect.[11,15] T_{cL} is seen to be a smooth function of the sheet resistance even going through the value corresponding to the threshold resistance for global superconductivity at low temperatures. Although T_{cL} is depressed when the sheet resistance is increased, the threshold condition is unaltered from that found when there was no significant depression, e.g., in the case of Sn films. Indeed, examination of previous work on a number of different thin film systems[13,14,16] reveals that regardless of the degree of depression of T_c, 6000 $\Omega/\square$ appears to be an upper limit for the threshold normal resistance.

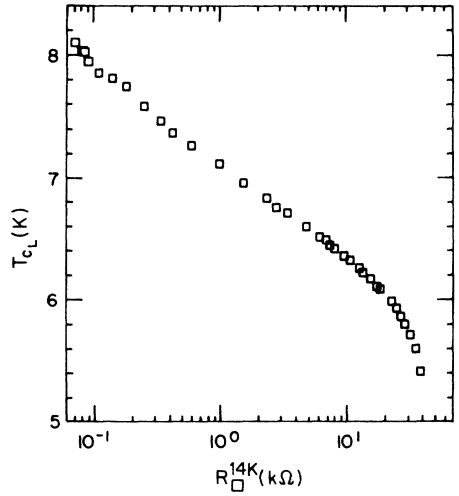

FIG. 3. The local transition temperature T_{cL} as defined by $R_\square(T_{cL}) = 0.5R_\square$ (14 K) vs normal-state resistance $R_\square$(14 K). The roll-off at high sheet resistances is a consequence of applying this definition of T_{cL} to traces of $R_\square(T)$ which exhibit a local minimum.

The models used to explain a resistance threshold actually do not address the issue of whether there is microscopic disorder within the clusters of a film which might reduce the magnitude of their transition temperature. The issues of the onset of zero electrical resistance and the depression of the transition temperature may have different physical origins. That is, although the depression of the transition temperature of a cluster depends upon the degree of microscopic disorder within it, as long as the latter does not completely depress local superconductivity, the condition for the occurrence of zero resistance at low temperature (global superconductivity) is determined only by the normal-state sheet resistance.

In summary, we have investigated the onset of superconductivity in ultrathin Ga films and found that it occurs only when the normal sheet resistance falls below a threshold which is close to $\pi/2(\hbar/e^2)$. This result confirms a conclusion drawn from previous measurements on Sn films and is consistent with measurements on other thin film systems. The fact that superconductivity has been taken to be the condition under which the resistance of the films is not measurable does not necessarily imply that a phase transition of the Kosterlitz-Thouless type[17] has actually taken place. Elucidation of the nature of the transition, if one actually occurs, would require extensive study of the I-V characteristics of the films which is currently underway and will be reported elsewhere.

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