SUPERCONDUCTIVITY OF SUPERIMPOSED METALS*

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Measurements[1-3] on thin films of superconductors in contact with normal metals have shown that the transition temperature of the films is lowered. For sufficiently thin films the transition temperature may be reduced to such an extent that superconductivity is not observed. There have been several theories of this effect.[1,4,5] Our experiments described here indicate that for films deposited by evaporation the effects observed can be considerably influenced by diffusion between the two metals.

We have made a series of measurements of the magnetic moment of superconducting tin films evaporated onto gold plates. The method used to detect the magnetic moment was similar to Alekseyevsky's,[6] in which the specimen is suspended from a torsion balance at a small angle to a uniform magnetic field. As the field strength is increased the film rotates until it is parallel to the field. The deflection is then constant until the critical field value is reached, when the specimen returns to its original position. This method is preferable to the measurement of resistance which has been used in all previous experiments. The resistance method has the disadvantage that it may not necessarily measure a bulk property of the film. Zero resistance may arise from superconducting filaments, or a finite resistance may result from small breaks in the film, neither of which is characteristic of the material as a whole. However, the magnetization is a bulk property, not affected by small discontinuities. Moreover, measurements of the magnetic moment as a function of temperature and magnetic field show whether or not the specimens behave like films of an ideal superconductor.

The substrates for our films were 99.99% pure gold plates 21 mm × 21 mm and of thickness 0.25 mm. The plates were annealed at 750°C for about a day and then wiped with trichlormethylene. During the evaporation they were firmly clamped to a hollow copper block cooled by flowing water at room temperature. The tin was evaporated from a molybdenum boat about 15 cm below the plate. During evaporation the pressure was less than 5×10^{-5} mm Hg. At the same time, the film was also deposited onto a thin glass cover slip which was next to the gold plate and symmetrically placed with respect to the boat. Between the boat and the plates was a shutter which was opened only after the tin had been melted, outgassed, and evaporation had begun. The shutter was kept open for 2.5 min for all evaporations so that every specimen saw the hot boat for the same length of time. The electrical power supplied to the boat also was the same for all depositions.

The anount of tin placed in the boat was weighed to give the thickness desired, and the evaporation time was sufficient to insure that all the tin vaporized. Thicknesses quoted in Table I were determined by weighing the glass slips before and after deposition. The films were squares of area about 3 cm².

Table I shows the results of the measurements on these films. There is a rapid decrease in transition temperature when the thickness of tin is less than about 7000 A; also, the films fall into two groups. Those specimens with transition temperatures close to the value of bulk tin (say $T_C > 3.4$°K) had sharply defined critical magnetic fields, and the magnetization and critical fields exhibited the behavior to be expected for thin films of an ideal superconductor. However, those films whose transition temperatures were appreciably depressed showed the characteristics of nonideal superconductors, having very high critical fields which were not sharply defined. Furthermore, their magnetic moments were smaller than those of the other films. The films deposited onto glass all had transition temperatures near the

Table I. Superconducting properties of tin films on gold.

Film	Thickness (A)	T_c (°K)	Behavior
5	18 700 ±500	3.71	ideal
8	13 200	3.40	ideal
9	9500	3.51	ideal
10	7800	2.68	nonideal
13	7100	3.57	ideal
14	7100	3.59	ideal
15	6100	3.08	nonideal
11	6100	1.66	nonideal
7	5700	not superconducting down to 1.35°K	
6	1500	not superconducting down to 1.35°K	
4	1500	not superconducting down to 1.35°K	

value of bulk tin.

These results appeared to confirm qualitatively the earlier observations of this effect. Misener and Wilhelm[1] found that tin films electroplated on constantan ceased to be superconducting above 2°K at thicknesses less than 3000 A. More recently, Meissner[2] measured several pairs of metals, including tin on gold, and reported a critical thickness of a few hundred angstroms. Smith, Shapiro, Miles, and Nicol[3] observed qualitatively similar effects with lead and silver films.

However, by further experiments we showed that under the conditions of our evaporations diffusion probably occurred despite the cooling precautions. This was established by replacing some of the specimens in the evaporator and exposing the films to the radiation from the hot empty boat, under conditions similar to those used in laying them down. After this treatment we measured their superconducting properties again and found them changed. The transition temperature of film 14 was reduced to 2.26°K after a 2.5-min exposure, and its magnetic behavior changed from ideal to nonideal in the sense already explained. After a further exposure of 2.5 min, film 14 was not superconducting down to 1.27°K. Film 9 after a 5-min exposure was not superconducting down to 1.23°K. The appearance of the films was unchanged after these exposures. Since the superconductivity of our films can be destroyed by heating the tin-gold surface, it follows that the value of the thickness at which the transition temperature is observed to fall rapidly has no particular significance, but is a consequence of the way the specimens were prepared.

We believe that the effects of reheating show that diffusion can easily occur during evaporation, and result in a decreased transition temperature. Furthermore, diffusion might account for the different values given by various workers for the thickness at which superconductivity disappears. The degree of diffusion and alloying which can take place will, of course, depend on the particular pair of metals used and the method of film deposition.

Gold is only soluble in tin up to 0.3% and about 4% of tin is soluble in gold, but intermetallic compounds $AuSn_4$ and $AuSn_2$ exist.[7] Allen[8] studied the superconductivity of tin-gold alloys and found that the transition temperature of the tin-rich alloys is independent of gold concentration up to about 30% gold. The intermetallic compounds are also superconducting, but alloys with more than 50% gold are not superconducting down to 1.5°K.

Although diffusion seems to reduce the transition temperature, the exact reason for this is not obvious. Appreciable interpenetration of the two metals could have two effects: Nonsuperconducting alloys may be formed; or the thickness of the superconducting metal could be reduced to a value much less than that deduced from the weight. Alternatively, there may be only a small interpenetration resulting in more perfect contact between the two metals,[9] which according to Cooper[5] can lower the transition temperature of relatively thick films.

In conclusion, we feel that we have shown that diffusion can play a critical role in experiments on the superconductivity of superimposed metals; and consequently, great caution must be used in the interpretation of such experiments until the role which diffusion plays is adequately understood.

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[9]We are indebted to Dr. Cooper for suggesting this possibility.