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Superconducting Behavior of Superimposed Thin Films of Al and Sn

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~~It is of interest to study the behavior of superimposed layers of superconducting and non-superconducting metals. We report ~~the~~ nuclear magnetic resonance (NMR) and resistivity measurements, on multiple alternating layers of Sn and Al in the temperature range between 1.5<sup>o</sup> K and 4.2<sup>o</sup> K. The accepted transition temperatures of Al and Sn are 1.2<sup>o</sup> K and 3.75<sup>o</sup> K, respectively.~~

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Others<sup>1</sup> have observed a depression of  $T_c$  in thin films of Sn or Pb when either is overlaid with Ag or Cu. A theoretical prediction<sup>2,3</sup> for film thickness less than the coherence length suggests that the specimen should exhibit a single depressed transition temperature characteristic of a diluted superconductor. It is tempting to regard Al above its transition temperature as the counterpart of the normal metal Ag, on which model one might expect a corresponding depression of the  $T_c$  of Sn in the Sn-Al combination. Our experimental results on Sn-Al sandwiches are that:  $T_c(\text{Sn}) = 3.78^\circ\text{K}$  with a mean deviation among 101 specimens of  $\pm 0.03^\circ\text{K}$ ; the extremal values were  $3.68^\circ\text{K}$  and  $3.82^\circ\text{K}$  and the width of any one transition was  $0.05^\circ\text{K}$  or less; and there is no evidence for a superconducting state in Al above  $1.76^\circ \pm 0.20^\circ\text{K}$ . These data are consistent with the tunneling experiments of Van Gorp<sup>4</sup> on similar samples.

Our films were prepared<sup>5</sup> by alternate evaporation of the separate metals in a vacuum of approximately  $10^{-6}$  mm Hg pressure onto a  $0.25 \times 10^{-3}$  inch mylar sheet, which was cooled to a temperature below  $0^\circ\text{C}$ . The thickness of the individual layers varied between 70 and  $320 \text{ \AA}$  for each metal; the total number of pairs varied from one to forty on each mylar sheet. For the measurement of resistivity a  $1 \times 3$  cm strip was cut from a mylar sheet, the usual four contacts being made to the metallic samples with silver paint. Approximately one thousand such strips were assembled to form a sample for NMR measurements.

TABLE I. NMR Samples

Sample	Number of pairs on mylar	Upper $T_c$ ( $^{\circ}\text{K}$ )	Lower $T_c$ ( $^{\circ}\text{K}$ )	$H_c(0)$ (kilogauss)	Sn film thickness	Al film thickness
1	40	3.78	(*)	4.0	160 $\text{\AA}$	70 $\text{\AA}$
2	1	3.73	1.68	8.5	160 $\text{\AA}$	120 $\text{\AA}$
3	5	3.76	(*)	7.5	160 $\text{\AA}$	240 $\text{\AA}$

(\*) Superconducting transitions at the lower temperature, identified as the Al transition, were obscured by a complete transition to zero resistivity at the upper  $T_c$  brought about by the high probability of having a conducting path of tin alone in these thicker multiple-layered samples.

NMR signals for  $\text{Al}^{27}$ ,  $\text{Sn}^{117}$ , and  $\text{Sn}^{119}$  in sample (1) identified resonances from the respective normal pure metals, resonance shifts, line widths and intensities suggesting no evidence for the formation of solid solutions or compounds of any kind.<sup>6</sup> Partial flux exclusion and the resulting internal magnetic field inhomogeneity prevented observation of the NMR in the superconducting state for this sample. In samples (2) and (3), however, the Al NMR was observed to have a width and resonance shift characteristic of normal Al down to 1.6 $^{\circ}\text{K}$  in a magnetic field of 4500 gauss (the amount of Sn in these samples was insufficient to provide observable signals at any temperature). Resistivity measurements of these samples

suggest that at least the Sn was superconducting below  $3.7^{\circ}\text{K}$ , and since neither the width nor resonance shift of  $\text{Al}$  changed, we interpret the measurements to mean that the  $\text{Al}$  was not superconducting. (Both aluminum<sup>6</sup> and tin<sup>7</sup> resonance frequencies are known to decrease slightly in the superconducting states in the pure materials.)

In order to interpret the resistivity measurements satisfactorily we must comment on the structure of the films. It is known<sup>7</sup> that Sn layers evaporated on mylar consist of platelets approximately  $100 \text{ \AA}$  in diameter, so that the films are not continuous. We find that a superimposed layer of  $\text{Al}$  restores the continuity. Furthermore, although an initial layer of  $\text{Al}$  is continuous, an overlayer of Sn reduces the conductivity. If the initial  $\text{Al}$  layer is thin, the overlayer of Sn results in a discontinuous film. Apparently local melting of the initial layer of Sn is produced by deposition of the second, and, since neither solid solutions nor intermetallic compounds exist, we visualize the sample as a conglomeration of small platelets of the separate metals in intimate physical contact. This interpretation is consistent with the measurements of resistivity. Of the 101 samples investigated some 13, primarily those containing the thinnest Sn layers, showed a sharp drop in resistivity at the Sn transition temperature to an intermediate value which remained constant down to approximately  $1.76^{\circ}\text{K}$ , below which the resistivity dropped to zero. The lower temperature varied at most by  $\pm 0.2^{\circ}\text{K}$  among the samples, but all the transitions were rather sharp having widths of  $0.05^{\circ}\text{K}$  or less. Of the other 85 samples all but five showed zero resistivity just below the upper temperature; in the five, a single transition to zero resistivity occurred at the lower temperature; in one or two cases no transition was observed.

In view of the likely platelet structure, we assume that the conducting path is divided between the two metals, the upper and lower transition temperatures being associated with Sn and Al, respectively. We note that Giaever and Megerle<sup>8</sup> also found enhanced  $T_c$  for Al up to 1.8°K for the thinnest films. It is important to emphasize that no transitions occurred between the two temperatures cited above. Whether or not the enhanced  $T_c$  in Al is to be attributed to an action of the Sn on Al or simply to strains in the latter, the fact remains that the NMR of the Al is typical of the normal metal even when it is proximate to Sn in the superconducting state.

We emphasize that our results pertain to a composite specimen the two components of which do not mutually dissolve or interact to form compounds. The same is not true of previously investigated specimens like Sn-Ag.<sup>4</sup> Our results suggest that the superconducting properties of the Sn component are not affected by intimate contact with a metal in the normal state. It is probably also true that the Al is not affected by the close proximity of superconducting Sn at least above the lower transition temperature.

For the sake of completeness it should be pointed out that all the thin film samples have characteristically high critical fields,  $H_c(0)$  ranging from 3000 to 11,000 gauss; this is convenient for the NMR measurements. Their critical fields also displayed either one of two characteristic angular dependences with respect to the magnetic field direction as discussed by Tinkham,<sup>9</sup> suggesting a first order transition for those with the lower critical fields, and a second order for those with the highest critical fields.



This work is being continued in order to investigate Cooper's suggestion that the NMR shift of both components may be affected when the entire specimen is superconducting.

We are grateful to Dr. R. H. Hammond of General Atomic, San Diego, for permission to quote his unpublished results on pure  $A^2$ . We acknowledge with thanks the assistance of W. Henry with some of the measurements. One of us (D.S.S.) wishes to express his appreciation to the Department of Physics of the University of California, Berkeley, for its hospitality during the course of this investigation.

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