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Tuning the transition temperature of superconducting

Ag/Pb films via the proximity effect

Physica C, **382**, 411, (2002).

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We report measurements of the transition temperature (T_C) of superconducting films composed of various combinations of Ag and Pb layers. For samples with good electrical contact between the layers, the measured T_C values show reasonable agreement with the Cooper model of the proximity effect. In poorly coupled samples, the normal layers appear to cause little if any suppression of the T_C . We present a simple predictive expression for T_C as a function of Ag content.

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The proximity effect occurs when a normal metal is placed in direct contact with a superconductor to produce well-defined phenomena [1-3]. The pair wavefunction of the superconductor tunnels into the normal metal thereby making it a superconductor. This spreading out of the superconductivity also dilutes it: both the transition temperature T_C and the superconducting energy gap Δ are reduced. In order to produce superconducting films with tunable T_C s between 4 and 7 K to act as screening ground planes [4], we have studied the proximity effect in Pb/Ag layers.

Pb and Ag films were thermally evaporated onto fire-polished glass substrates held at room temperature. A quartz crystal microbalance was used to measure the average thickness of each deposition. These samples were then mounted in a low temperature probe which was evacuated and submerged in liquid helium. A Ru-oxide thermometer and a resistance heater were used to measure and control the temperature respectively. Sample resistance was measured using a standard *4-wire* technique. Current vs. voltage (I - V) curves were produced at various fixed temperatures in order to determine the T_C of each sample. Since these samples were relatively thick (order *50-100 nm*), transitions appeared to be very abrupt, leading to easily determined T_C values.

A subset of the samples were made by depositing either the Pb or the Ag and then breaking vacuum for about 1 minute to change the evaporator source. During this procedure, dry nitrogen gas was passed through the chamber in order to reduce oxidation of the surface. The relevant parameters for these samples are shown in table 1. Note that Ag amounts are divided into underlayer and overlayer thicknesses denoting Ag layers

deposited before and after the Pb respectively. The remainder of the samples were deposited using a 2-source evaporator which allowed the constituents to be changed without breaking vacuum. Parameters for these samples are presented in table 2.

We compare our results for T_C with both the Cooper model for the proximity effect [1] and a simple linear recipe. In the former prediction, the interaction constant from the Cooper pair model, $[N(0)V]_{1+2}$ in a bi-layer sample of superconductor of thickness d_1 , and a normal metal of thickness d_2 is given by:

$$[N(0)V]_{1+2} = \frac{d_1}{d_1 + d_2} [N(0)V]_1.$$

It should be noted that a corrected expression as given by de Gennes [2] also includes the weights of the Fermi level density of states of each material. The simpler Cooper expression that we use is truly valid only if the density of states of the two materials is the same. According to the BCS result for the transition temperature

$$k_B T_C = 1.13 \hbar \omega_c e^{\frac{-1}{N(0)V}},$$

so that the sample transition temperature, $T_{C(Pb+Ag)}$, is given by

$$\frac{T_{C(Pb+Ag)}}{T_{C(Pb)}} = \frac{e^{\frac{-1}{[N(0)V]_{Pb+Ag}}}}{e^{\frac{-1}{[N(0)V]_{Pb}}}} = e^{\frac{-d_{Ag}}{d_{Pb}}}.$$

In this expression $T_{C(Pb)}$ is the transition temperature of the pure superconductor, in our case Pb at 7.2 K. We have plotted this prediction as a solid curve in Figure 1. We have chosen as our abscissa the thickness fraction of Ag:

$$\frac{d_{Ag}}{d_{Pb} + d_{Ag}}.$$

Based on the results from quench condensed Ag/Pb films [5] and the linear scaling of the interaction constant, we have also included a linear recipe for T_C suppression as a function of Ag thickness fraction (plotted as a dotted line), where

$$T_{C(Pb+Ag)} = T_{C(Pb)} \left(1 - \frac{d_{Ag}}{d_{Pb} + d_{Ag}} \right) = T_{C(Pb)} \left(\frac{d_{Pb}}{d_{Pb} + d_{Ag}} \right).$$

Note that there is a relatively small difference between this linear recipe and the Cooper model within our temperature range.

The T_C for each sample included in Tables 1 and 2 is plotted vs. thickness fraction of Ag in Figure 1. In films that were produced by depositing Pb first, then breaking vacuum before depositing Ag, there was no obvious T_C suppression. T_C values for these samples are not shown (indistinguishable from the T_C of pure Pb). Films produced by depositing Ag first, then breaking vacuum before depositing Pb (filled diamonds, samples $1n$ through $9n$), *did* show proximity effect reduction of the T_C . However, if the vacuum was broken a second time and subsequent Ag was deposited, the T_C data showed poor correlation with the total Ag thickness (open circles, samples $10n$ through $23n$). Our preliminary hypothesis is that the Ag does not oxidize substantially when the vacuum is broken, but the Pb does. If an oxide weakens the coupling between the sample layers, the proximity effect is reduced. Following this assumption, we have also plotted the data for samples $10n$ through $23n$ using only the underlayer Ag in the thickness fraction

calculation (closed circles). Note the better agreement between the data and the predictions.

In order to further test our hypothesis, we configured a 2-source evaporator so that Ag could be deposited onto Pb without a break in vacuum. Results for these samples are also shown in Figure 1. Samples with an Ag underlayer only (1v through 6v) are indicated by closed squares. For the tri-layer samples (7v through 14v) we plot the measured T_C versus both the underlayer Ag thickness fraction (open triangles) as well as the total Ag thickness fraction (closed triangles). In these samples, using *only* the underlayer thickness appears to predict a larger suppression of the T_C than our overall trend. In fact, we believe that the relatively good vacuum conditions used in producing these samples require that the total Ag thickness fraction be used as the relevant parameter for predicting the T_C . Our conclusion is that we should include all Ag which is in good electrical contact with the Pb. Any material deposited over Pb after a break in vacuum apparently does not significantly contribute to the proximity effect.

Our results are consistent with Cooper's modified reduction factor

$$\frac{d_{Pb}}{d_{Pb} + \beta d_{Ag}},$$

where β is due to any barrier between the layers [1]. Since Pb should be expected to oxidize more readily than Ag, the value for β should be much closer to unity for barriers over Ag when compared to barriers over Pb. In the samples prepared without breaking vacuum, we expect values of β close to unity for all layers.

We conclude that only Ag which is in good electrical contact with the Pb produces significant T_C suppression. It appears as if breaking vacuum causes an electrical barrier to form on Pb which inhibits proximity coupling, whereas the maintaining of the vacuum has no such occurrence. This barrier does not form as readily on the Ag, so samples formed by depositing Ag first and breaking vacuum before the Pb layer *do* show proximity effects. Using this assumption, we have plotted all “good contact” data with closed symbols. Although the data follow a consistent trend, there is still significant scatter in our results beyond the expected random error. This variation is perhaps explained by differences in evaporation parameters such as vacuum quality and evaporation rate which might affect changes in film purity and morphology. For our regime of interest, we have adopted a linear recipe for determining relative Pb/Ag thicknesses in order to tune our samples to the desired T_C . This recipe does not differ significantly from the Cooper prediction, and certainly varies by an amount much less than our experimental scatter. Since our samples are only of order *100 nm* thick, we cannot assume that this recipe works for a wide range of sample thicknesses. It is certainly consistent with previous results on quench condensed films [5, 6]. However, recent work on very dilute proximity effect samples does indicate better agreement with the Cooper model [7].

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Captions

Table 1: Results for Pb/Ag films with nitrogen venting between depositions. The measured transition temperature is compared to the thickness fraction of Ag in each sample. The solid symbols denote thickness fractions of the Ag which is in good electrical contact with the Pb (Ag layers under the Pb layers). The open circle denotes total Ag thickness fraction.

Table 2: Results for Pb/Ag films deposited under good vacuum conditions. The measured transition temperature is compared to the thickness fraction of Ag in each sample. The solid symbols denote thickness fractions of the Ag which is in good electrical contact with the Pb (in this case the total Ag thickness). The open triangles denote Ag thickness fractions for only the underlayer Ag.

Figure 1: Transition temperature vs. thickness fraction for the samples listed in Tables 1 and 2, the prediction of the Cooper model, and our linear recipe. The solid symbols represent Ag thickness fractions calculated using only the Ag layers in good contact with the samples. The solid curve shows the Cooper Model and the dashed line represents the simple linear recipe for predicting sample transition temperatures.

Sample	Ag under (nm)	Pb (nm)	Ag over (nm)	Ag total (nm)	Ag total fraction	Ag under fraction	Ag total fraction	T _C (K)
					bi-layer	tri-layer	tri-layer	
					◆	●	○	
1n	0	71.5	0	0	0			6.90
2n	0	99.2	0	0	0			7.15
3n	0	51.5	0	0	0			7.12
4n	7.5	45.1	0	7.5	0.14			6.48
5n	11.9	52.8	0	11.9	0.18			6.10
6n	11.9	48.4	0	11.9	0.20			6.18
7n	14.3	47.9	0	14.3	0.23			5.94
8n	14.3	47.3	0	14.3	0.23			6.04
9n	14.0	29.7	0	14.0	0.32			5.84
10n	8.4	74.6	13.4	21.8		0.10	0.23	6.48
11n	10.3	80.3	12.5	22.8		0.11	0.22	6.80
12n	9.6	71.8	11.9	21.5		0.12	0.23	6.70
13n	9.9	75.4	12.3	22.2		0.12	0.23	6.33
14n	7.2	48.4	11.9	19.1		0.13	0.28	6.30
15n	10.5	67.1	16.7	27.2		0.14	0.29	5.79
16n	10.0	60.0	16.0	26.0		0.14	0.30	6.04
17n	8.8	49.5	14.3	23.1		0.15	0.32	5.65
18n	10.0	52.8	13.4	23.4		0.16	0.31	5.91
19n	10.7	55.4	14.7	25.4		0.16	0.31	5.56
20n	9.3	44.3	12.7	22.0		0.17	0.33	5.49
21n	9.4	46.9	13.1	22.5		0.17	0.33	6.33
22n	9.6	44.7	13.7	23.3		0.18	0.34	5.62
23n	11.9	31.9	9.6	21.5		0.27	0.40	5.39

Table 1

Sample	Ag under (nm)	Pb (nm)	Ag over (nm)	Ag total (nm)	Ag total fraction	Ag under fraction	Ag total fraction	T _C (K)
					bi-layer	tri-layer	tri-layer	
					■	△	▲	
1v	15.3	87.8	0	15.3	0.15			6.03
2v	21.1	88.1	0	21.1	0.19			5.96
3v	16.0	66.0	0	16.0	0.20			5.94
4v	23.2	89.1	0	23.2	0.21			5.95
5v	35.8	88.1	0	35.8	0.29			5.96
6v	19.9	37.3	0	19.9	0.35			4.95
7v	6.8	58.0	12.1	18.9		0.10	0.25	5.34
8v	7.4	44.3	12.0	19.4		0.14	0.30	5.12
9v	10.2	44.8	11.4	21.6		0.19	0.33	5.21
10v	5.6	27.2	8.4	14.0		0.17	0.34	4.86
11v	13.4	44.3	12.0	25.4		0.23	0.36	4.85
12v	14.9	44.5	12.1	27.0		0.25	0.38	4.60
13v	12.3	44.2	16.5	28.8		0.22	0.39	4.26
14v	15.9	44.0	11.9	27.8		0.27	0.39	4.90

Table 2

