Tunnel-Effect Measurements on Superimposed Layers of Lead and Aluminum*

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Superimposed Al and Pb films are studied using the tunnel effect as an indication of the influence of a normal metal (Al) on a superconductor (Pb). The structures are composed of four layers, $Al/Al_2O_3/Pb/Al$. The Pb and top Al layers were vacuum-evaporated onto 77°K substrates to eliminate diffusion problems. Curves are presented which show the effect of an overlayer on the tunnel-junction *I-V* characteristic and density-of-states function. The film thicknesses are varied over a wide range. The variation in energy gap with Pb thickness is plotted for a constant Al overlayer of 3100 Å. In addition there are three samples made to determine the critical temperature by resistive measurements with the same structure as a check on the reliability of the tunnel-function results. The energy-gap transition occurs between 300 and 4500 Å of Pb. Comparison with Hauser and Theuerer's results on superimposed Al and Pb films is included in the discussion.

INTRODUCTION

It has been shown by Giaever that an approximation of the density-of-states function of a superconductor can be experimentally determined through the use of superconducting tunnel junctions.[1,2] This paper reports the use of the same technique to observe the narrowing energy gap of a thin film of Pb when overlaid with Al. The change in the density of states function of a thin film of superconducting Pb overlaid with Al was observed as the Pb thickness was varied. Overlayer experiments involving Pb and Al have previously been undertaken by Hauser and Theuerer using a resistive sample whose Pb layer was sputtered at low temperatures.[3,4,4a] Resistive samples were made also in this laboratory to check the results of the tunnel devices.

SAMPLE PREPARATION AND TESTING

All samples were of a four-layer structure vacuum-deposited on glass substrates measuring $\frac{3}{8}$ in.$\times\frac{1}{4}$ in. First, about 1000 Å of 99.99% Al was deposited on the glass substrate and allowed to oxidize in air for 15 min. Then varying amounts of Pb and Al were deposited in a strip crossing the $Al+Al_2O_3$ layers to form a tunnel junction of approximately 1 mm^2. The final two layers were deposited in rapid sequence on substrates which were cooled to 77°K just pripor to evaporation. The samples were kept at temperatures below 100°K at all times until testing was completed. Lead wires were soldered with indium to the glass substrates before evaporation. This procedure eliminates diffusion problems and also minimizes the possibility of interlayer contaminants. The top Pb and Al layers were deposited throughout the same mask since the tunnel current is a distributed current and edge effects should be relatively unimportant. Samples tested at 4.2°K were immersed in the liquid-He reservoir after threading the samples and lead wires through a tubular stainless steel sample holder. This eliminated the necessity to transfer liquid He every time a sample was tested at this temperature. Current-voltage (*I-V*) characteristics were plotted directly on a Mosley 1030-A *X-Y* plotter and the current was normalized at 5 mV. Three identical samples were evaporated at the same time as a consistency check. One of the three was covered during the final Al evaporation so that a check was obtained on the basic $Al/Al_2O_3/Pb$ part of the tunnel junction. Thickness measurements were all obtained by the Tolansky method and are accurate within ±10%.

EXPERIMENTAL RESULTS

Experimental data are first presented in the form of a family of curves (Figs. 1 and 2) from different samples covering the range of thicknesses where the transition occurs. The temperature is held constant at 4.2°K. The

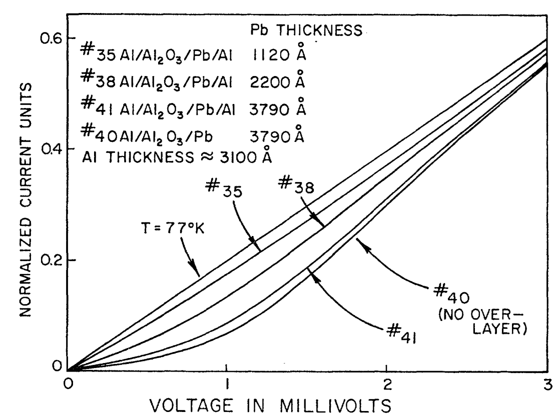

FIG. 1. Current-voltage characteristics of tunnel junctions at 4.2°K. Al overlayer held constant at 3100 Å for all samples except #40 which has no overlayer. Pb thickness is varied. The straight line is the common normalized characteristic for all samples at 77°K.

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[1] I. Giaever, Phys. Rev. Letters **5**, 147 (1960).
[2] I. Giaever and K. Megerlie, Phys. Rev. **122**, 1101 (1961).
[3] J. J. Hauser, H. C. Theuerer, and N. R. Werthamer, Phys. Rev. **136**, A637 (1964).
[4] J. J. Hauser and H. C. Theuerer, Phys. Letters **14**, 270 (1965).
[4a] *Note added in proof.* See also G. Bergmann, Z. Physik **187**, 395 (1965).

Al overlayer thickness is held constant at 3100 Å in Fig. 1. Sample 40 is the basic $Al/Al_2O_3/Pb$ tunnel junction. It represents the case where the Pb thickness is very large and any Al overlayer would have no effect on the characteristic. Sample 41 shows that the overlayer starts to affect the characteristic when the Pb is around 3790 Å thick. Even for sample 35 there is a retardation of electron flow across the barrier suggesting an energy gap or at least a lower probability of tunneling around the Fermi level. The Pb transition appears to occur, therefore, between 500 and 5000 Å for an Al overlayer of 3100 Å.

Figure 2 shows results on samples with a constant Pb thickness of 2200 Å. Sample 31 has no Al overlayer, showing the full tunnel characteristic. With the addition of 2860 Å of Al, sample 38 gives about half the total superconducting current depletion of the basic junction. However, one can not tell from this figure that the I-V characteristic for a sample with 2200 Å of Pb and a very thick Al overlayer would go to a straight line without indication of superconductivity at 4.2°K.

Thermal "spreading" of the electron densities around the Fermi level causes the I-V characteristic of a tunnel junction to be strongly dependent on temperature. Figure 3 indicates that as the temperature is lowered through 1.22°K, the I-V characteristic appears to approach the ideal case at $T=0°K$ for which there is no current flow below $V=\epsilon$, where 2ϵ is the energy gap expressed in eV. Samples 34 and 35 were made simultaneously, except 34 was shielded from the final Al evaporation. As the temperature is lowered, there appears a more striking difference in the I-V characteristic.

It has been shown by several authors[1,5] that the normalized derivative of the current with respect to the voltage $[(dI/dV)/(dI_n/dV)]$ in a tunnel junction is an approximate representation of the density of states function. dI/dV is the slope of the I-V characteristic and dI_n/dV is the slope of the characteristic at 77°K

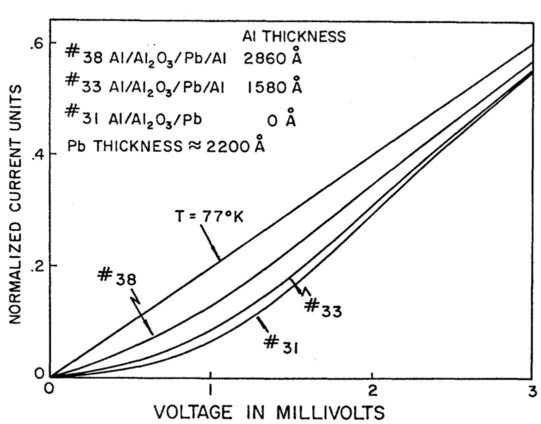

FIG. 2. Current-voltage characteristics of tunnel junctions at 4.2°K. Pb layer held constant at 2200 Å for all samples. Al thickness is varied.

[5] S. Shapiro, P. H. Smith, L. Nicol, J. E. Miles, and P. F. Strong, IBM J. Res. Develop. 6, 34 (1962).

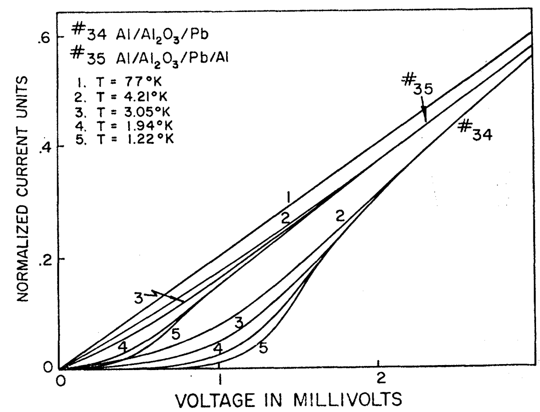

FIG. 3. Current-voltage characteristics of tunnel junctions at various temperatures. #34 and #35 were both made at the same time, #34 being shielded from the Al overlayer evaporation. Pb thickness is 1120 Å for both samples. Al overlayer thickness is 3240 Å.

(a constant). This representation becomes more accurate as the temperature approaches 0°K. Figure 4 is a plot of $[(dI/dV)/(dI_n/dV)]$ versus V, taken from selected curves of Fig. 3. It now becomes clear that the overlaid junction has a density of states function which is very similar to that of the ordinary junction except that the overlayer reduces the energy gap $\epsilon(T)$ of the Pb.

As T approaches 0°K, the point in Fig. 4 where $[(dI/dV)/(dI_n/dV)]=1$ would correspond to ϵ. For several samples we have measured the I-V characteristic as a function of temperature and specified the voltage for each temperature where

$$[(dI/dV)/(dI_n/dV)]=1$$

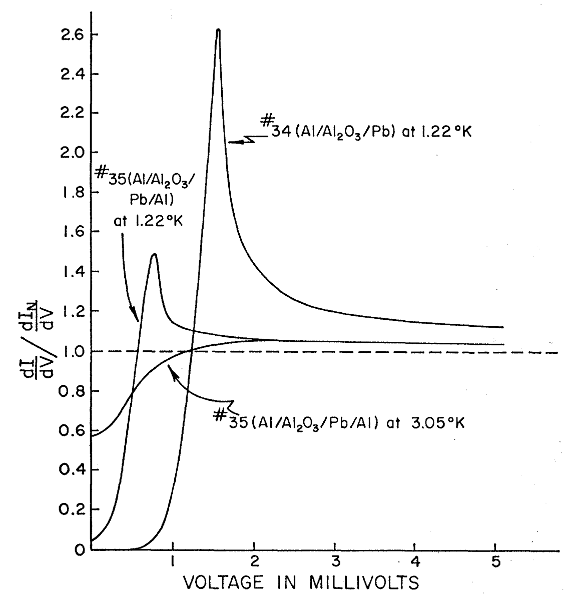

FIG. 4. $(dI/dV)/(dI_n/dV)$ versus V approximates the density-of-states function for a sample as $T\rightarrow 0°K$. Data taken from Fig. 3. Pb thickness is 1120 Å. Addition of 3240 Å of Al to sample #35 narrows the energy gap.

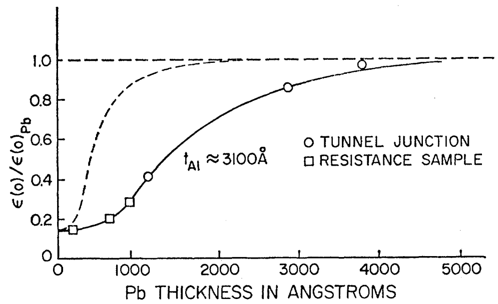

FIG. 5. Relative gap width versus Pb thickness for a heavy Al overlayer (3100 Å). The dotted curve is Hauser and Theuerer's result for sputtered films.

to be δ. Clearly, as $T \to 0$, $\delta \to \epsilon(0)$, where $\epsilon(0)$ is the energy gap at $T=0$°K. We have therefore obtained approximate values of ϵ for each sample by constructing the δ-versus-T plot and extrapolating at 0°K. For the samples recorded, δ varies quite slowly below 2°K and hence the extrapolated values are very close to the values at 1.22°K. Figure 5 shows the results of this procedure for a constant Al overlayer of 3100 Å.

In order to check the results of the tunnel junctions, resistive samples were made using the same 4-layered structure and the critical temperatures (T_c) were recorded. On the assumption that $\epsilon(0)$ is proportional to T_c, the data for three samples are recorded also in Fig. 5, where the vertical axis can be considered either $\epsilon(0)/\epsilon(0)_{Pb}$ or T_c/T_{cPb}. The intercept on the vertical axis corresponds to the critical temperature of the Al overlayer. The residual resistance of this type of sample is primarily the resistance of the Al layer as is indicated in Hauser and Theuerer's article on sputtered films. This was taken into account when determining the T_c of the samples as suggested by Milligan.[6] The resistivity of the evaporated Al films at 4.2°K was $6.83\times10^{-7}\,\Omega$ cm.

DISCUSSION

Since all work on the superimposed layers was done at 100°K or lower, the usual problems of diffusion can be disregarded. Samples were stored in liquid nitrogen and the tunnel resistance and the normalized tunnel characteristics remained unchanged after a month of storage.

The reliability of the observed phenomenon was borne out by the consistency of the many samples manufactured. There were no overlaid samples with I-V characteristics which did not fit the general pattern of results. When the normalized characteristics of two Al/Al_2O_3/Pb/Al samples with the same layer thicknesses were compared, the point by point resistances were equal to within $\pm2\%$. When the normalized characteristics of any two Al/Al_2O_3/Pb samples with different Pb thicknesses (350 to 3790 Å) were compared, the point-by-point resistances were also equal to within $\pm2\%$. These tolerances would be impossible to attain if the effects were significantly influenced by such random determinants as evaporation contamination, edge effects, gaseous interlayers, or dirty substrates.

It is significant that it is possible to obtain the same results with either the tunnel method or resistance method, as illustrated in Fig. 5. This supports the assumptions made in obtaining the Pb energy gap from the slope of the I-V characteristics. These results are somewhat different from the results of a sample with the glass/Al/Pb structure as reported by Hauser and Theuerer on sputtered films. Their samples yield a transition from $\epsilon(T)_{Al}$ to $\epsilon(T)_{Pb}$ in the range from about 100 to 1500 Å of Pb.[4] A sample of the type glass/Pb/Al with a Pb thickness of 600 Å and an Al thickness of 3000 Å was evaporated onto a 77°K substrate in this laboratory. The critical temperature for this sample was above 4.2°K in agreement with Hauser and Theuerer's work. At the time measurements were made, a cryostat for temperatures above 4.2°K was not available. On the other hand, when the superimposed layers are of the form glass/Al/Al_2O_3/Pb/Al, the Al seems to have a more profound effect on the Pb layer, causing the transition to occur between 300 and 4500 Å, as indicated by Fig. 5. The difference in the results at first notice might be attributed to the tunnel junction having an Al layer on both sides of the Pb layer, separated slightly (20–30 Å) on one side by an Al_2O_3 interlayer. This is not obvious, however, since as reported earlier the Pb layer of an Al/Al_2O_3/Pb tunnel junction is unaffected by the bottom Al layer at least down to 350 Å of Pb. The difference between the sputtered and the evaporated sandwiches cannot be completely explained as we did not make sputtered samples at this time. Perhaps the most significant difference in the samples is that in one case the critical Pb layer is condensed on an Al_2O_3 substrate. In the other it is condensed on either an Al or a glass substrate. Hilsch has shown that the results are shifted similarly on films of superimposed Pb and Cu when the evaporation sequence is reversed.[7] He suggests that the germination of the condensed layers on different substrates leads to different electron mean free paths. The fact that the Al has a stronger influence on the Pb films in these experiments leads one to believe that the electron pairs can more readily flow into the Al film and that impurities and interlayers are at a minimum.

Other work in this laboratory on Pb and Al tunnel junctions includes a report by Gross[8] on Al/Al_2O_3/Al/Pb structures where the top Al layer (between Al_2O_3 and Pb) was made thin enough to display superconducting characteristics at 4.2°K due to the superimposed Pb layer. Gross found that there was a current depletion at low voltages (similar to Fig. 1) in the tunnel junction I-V characteristic at 4.2°K for Al interlayers below 330 Å thick when the Pb overlayer was about 1000 Å. These structures were also vacuum evaporated at 77°K in order to minimize diffusion.

[6] E. J. Milligan, Cryogenics 5, 49 (1965).

[7] P. Hilsch and R. Hilsch, Z. Physik 180, 10 (1964).

[8] Thesis for the M. S. degree in Electrical Engineering by M. A. Gross, presented in 1964 (unpublished).