Superconductivity:
Percolation in Lead Films

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Abstract: Thin lead films with silicon encapsulation were made by evaporation onto liquid nitrogen-cooled silicon substrates under vacuum in order to study percolation. Films with average thicknesses of 575 Å, 150 Å, 100 Å, and 50 Å of lead were produced, and their penetration depths were taken. Critical temperature and superfluid density decreased as lead thickness decreased, and the penetration depths exhibited no frequency dependence. Near the superconducting-to-normal transition at T = Tc, the two-dimensional magnetic penetration depth $\lambda_\perp^{-1}$ for the 575 Å film was proportional to $(1\text{-}T/T_c)^{5/6}$ while $\lambda_\perp^{-1}$ was proportional to $(1\text{-}T/T_c)^{5/8}$ or $(1\text{-}T/T_c)^{2/3}$ for 150 Å and 100 Å films. The 50 Å film was discontinuous. The decreasing exponent likely indicates proximity to a percolation threshold between 50 Å and 100 Å thick.

Background:

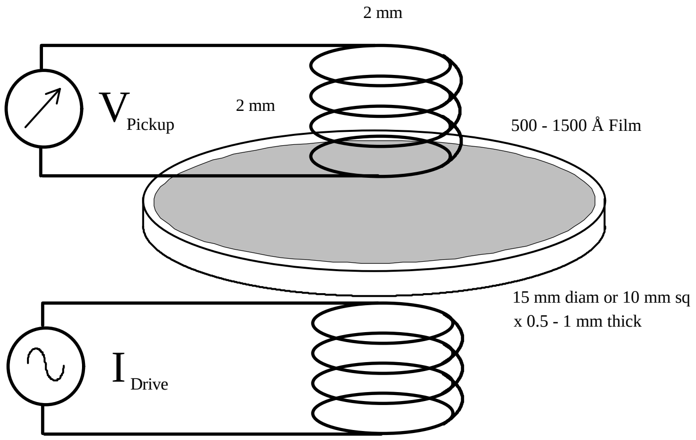
A superconductor has no resistance below a critical temperature T_c and, unlike a perfect conductor, always possesses no internal magnetic flux. A perfect conductor when cooled below its transition temperature will work to maintain the internal magnetic flux it possessed while passing through T_c. If the external field is varied below the transition, eddy currents are generated so that the critical temperature flux is maintained. However, the flux through a superconductor will always be zero below T_c. This is known as the Meissner Effect. If a superconductor with a nonzero flux is cooled below its transition temperature, it will generate eddy currents in a direction and magnitude such that the magnetic field produced by the currents will exactly cancel the external magnetic field.

If the external field is adjusted while the superconductor is below T_c, the magnitude and direction of the eddy currents will also shift so that the absence of internal flux is maintained.

Theoretically, the flux should fall abruptly to zero at the surface of the superconductor, but because the currents generated at the surface occupy a volume, it is impossible for the flux to cease precisely at the boundary. Instead, the flux decreases exponentially as it enters the superconductor. The distance in which the flux drops to 1/e of its surface value is known as the penetration depth. The penetration depth and the critical temperature are used most frequently to characterize a superconductor.

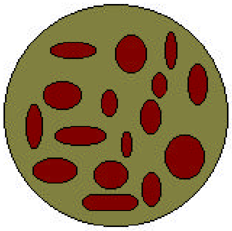
In order to measure the penetration depth λ, a thin film is placed between two coils as in Figure 1, and the mutual inductance of the coils is measured as a function of temperature. To measure the mutual inductance, a magnetic field is generated by sending current through the drive coil. If the film is above T_c, the magnetic flux generated by the drive coil passes through the film and causes a change in the magnetic flux through the pick-up coil. The changing magnetic field through the pick-up coil induces an EMF, which can be measured. Below T_c, the film allows a much smaller portion of the magnetic flux through it and thus partially shields the pick-up coil from the drive coil. The mutual inductance measurements are used to calculate the superfluid density, which is inversely proportional to the penetration depth. The superfluid density is the number of superconducting electron pairs, or Cooper pairs, per unit volume. The superfluid density increases as a power of $(T_c - T)$ as the temperature is decreased below the critical temperature, T_c.

FIGURE 1: set-up for mutual inductance measurements of a film



A percolating film is one which grows as islands that gradually merge together (Figure 2). This occurs because the metal atoms are more attracted to one another than to the substrate, and thus the atoms tend to bead up, leaving channels of substrate between islands of superconducting atoms. If the film is thin enough, the diameters of the islands are small enough so that the islands do not join, leaving the film discontinuous.

Figure 2: Top View of Islands of lead (red)
separated by silicon substrate (green)
before the islands are large enough to merge



As the film is cooled through its critical temperature, the islands become superconducting first, and then the channels between them gain superconductivity. When the channels become superconducting, the islands are connected to one another and their individual phases have approximately the same value over the entire surface. This enables the electron pairs to move freely over the surface of the film. Due to the dual superconducting transition in a percolating film, the onset of phase coherence occurs over a wider temperature interval than in a continuous film, and this is shown in the superfluid density maintaining a power law relationship over a larger temperature range near T_c for the percolating film.

The onset of phase coherence can be studied by looking at the two-dimensional magnetic penetration depth, $\lambda_\perp$, where $\lambda_\perp^{-1}(T) \equiv d/\ \lambda^2(T)$ and d is the thickness of the film. For a continuous phase transition, $\lambda_\perp^{-1}$ rises from zero at the transition temperature at a rate proportional to $(1-T/T_c)^x$. For a uniformly thick film, $x = 1$. The value of x is expected to decrease as the thickness of the film approaches the percolation threshold because the temperature range for the phase transition is greater than that of a completely continuous film, due to the need to connect the islands of superconducting atoms. Below the percolation threshold, the film is so discontinuous that a phase transition cannot occur.

Experimental Set-up:

In order to look closely at the percolation effect, lead films of various thicknesses were deposited on silicon substrates using a coevaporator system (Figure 3). The depositions occurred under vacuum in order to minimize impurities in the films, and the typical pressure within the bell jar (Figure 4) was on the order of 10^{-6} Torr. To evaporate the films, lead shot was placed in an evaporation boat (Figure 5) inside the bell jar. After sufficient vacuum was achieved, about 80 A of current was run through the boat in order to melt the lead. Some of the faster moving lead atoms evaporated and deposited on the silicon substrate hanging above the evaporation boat. The substrate was attached to the bottom of a copper block with vacuum grease, and the thickness of the film was measured by a thickness monitor.

Figure 3: The Coevaporator System

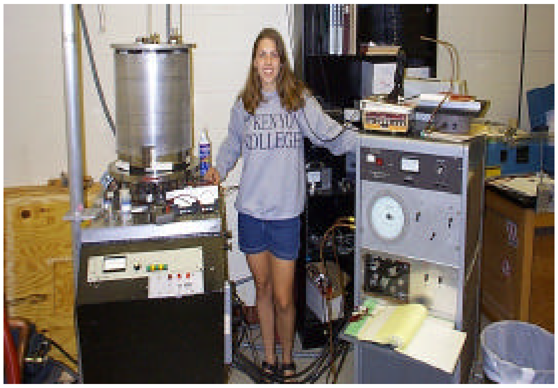


Figure 4: Inside Bell Jar
Top View

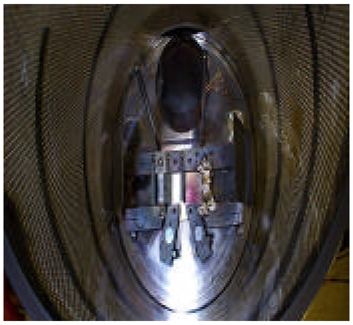
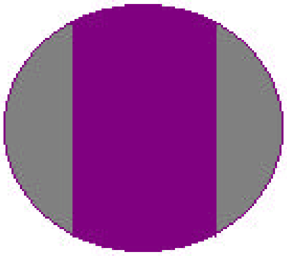


Figure 5: Evaporation Boats
For lead (right) and aluminum

To minimize the migration of lead atoms, liquid nitrogen was run through the substrate holder during the deposition. Silicon was then evaporated over the lead in order to encapsulate it, locking the film pattern into place. The silicon was evaporated by running 200 A of current through a tantalum boat, and it was assumed that the heat generated by this evaporation did not affect the lead because the substrate was cooled by liquid nitrogen.

Essentially, the addition of a lead layer is equivalent to putting a resistance in parallel with the silicon substrate. (The silicon was doped and therefore had its own electrical conductivity.) In order to measure the resistance of the film during the lead deposition, silver edges were evaporated onto the silicon substrate to establish electrical contacts (Figure 6), with the rest of the substrate being protected by an aluminum foil mask. The edges of the foil mask (the parts not touching the substrate) were attached to the substrate holder with vacuum grease. A feedthrough composed of copper wires run through a rubber stopper was made, and it was found to hold vacuum. The copper wires inside the bell jar were attached to the silver edges of the substrate with indium pads, and since the copper wires extended outside the bell jar through the rubber stopper, the resistances of the films were easily measured during the deposition. In this way, lead films of desired resistances could be produced.

Figure 6: Silicon substrate
with silver edges

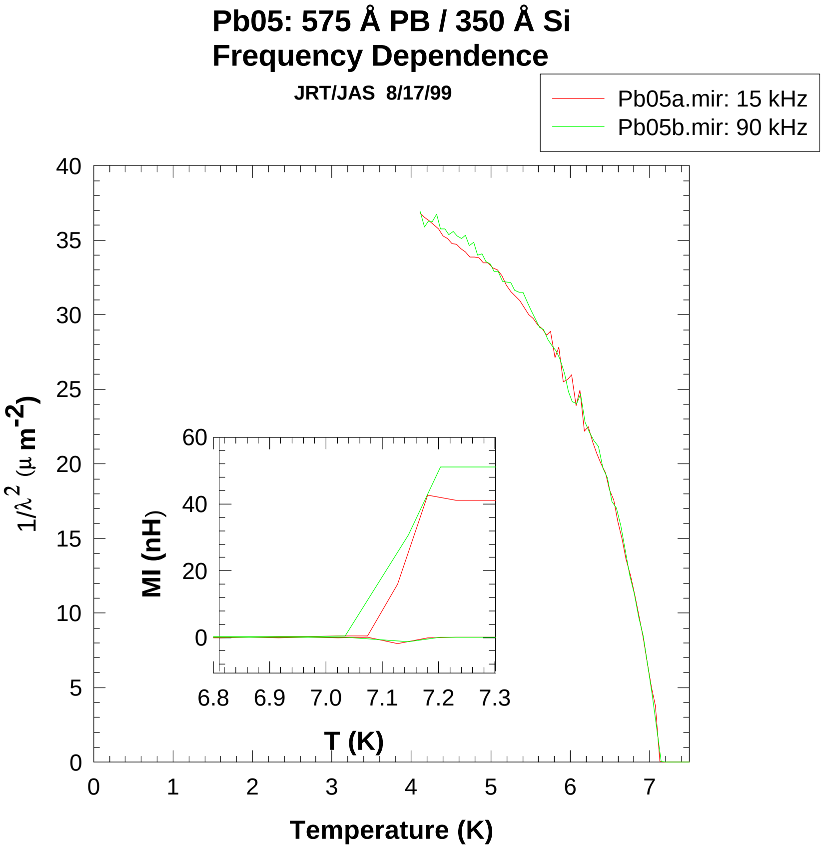


The penetration depths of the films were measured at 15 kHz and 90 kHz in order to determine if there is any frequency dependence in the superfluid density.

Results:

A thick lead film was grown (575 Å with 350 Å of silicon on top), and its penetration depth was taken in order to compare thinner films with one that is certainly continuous and whose thickness is more nearly uniform (Figure 7).

Figure 7: 575 Å Lead / 350 Å Si
$1/\lambda^2 \propto$ Superfluid Density
MI = mutual inductance



For the 150 Å lead film coated with 325 Å of silicon, the superfluid density decreased smoothly to zero as the temperature increased toward the transition, and the power law relationship did not maintain over an obviously wide temperature interval near the transition. Thus it was concluded that the percolation threshold was not achieved (Figure 8). Also, the superfluid density did not exhibit significant frequency dependence near the critical temperature. The 100 Å lead film coated with 90 Å of silicon was also above the percolation threshold, and no frequency dependence was observed (Figure 9).

Figure 8: 150 Å Pb/ 325 Å Si
$1/\lambda^2$ α Superfluid Density
MI = mutual inductance

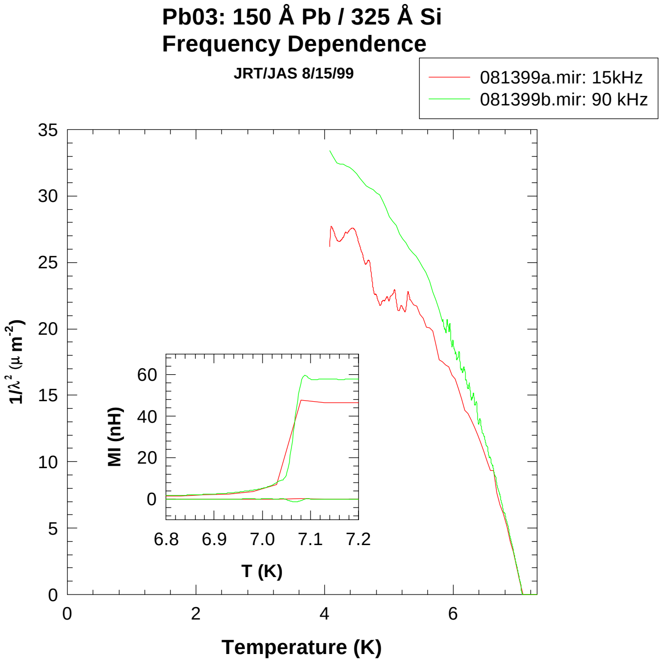
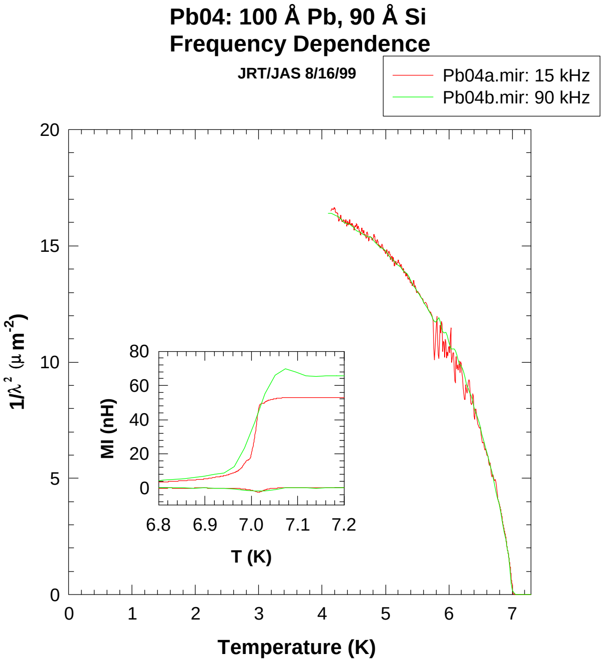


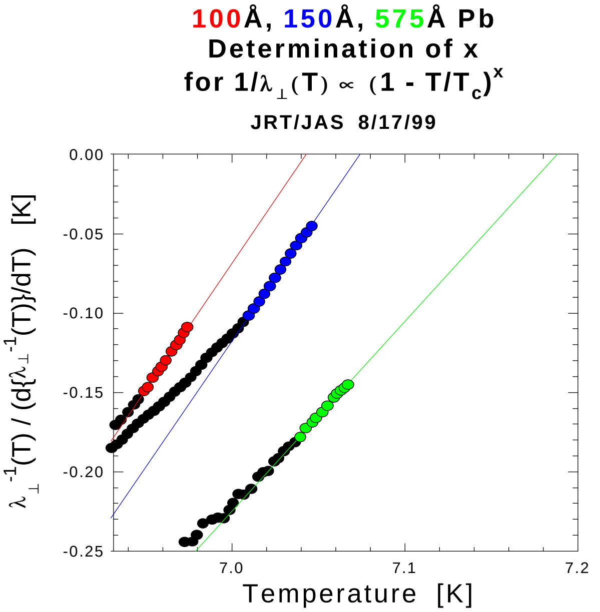
Figure 9: 100 Å Pb/ 90 Å Si
$1/\lambda^2$ α Superfluid Density
MI = mutual inductance



From the comparison to the bulk lead film, it is clear that the 150 Å and 100 Å lead films are continuous, since both films maintained their power law relation for almost the same temperature interval as the thick lead film. There was also no observable frequency dependence in the thinner films. It is important to notice, however, that the transition temperature decreases as the thickness of the lead decreases. This can be best seen on the mutual inductance versus temperature insets in Figures 7-9. (The out-of – phase peaks show this clearly.) The transition temperatures are approximately 7.14 K for the 575 Å film, 7.07 K for the 150 Å film, and 7.02 K for the 100 Å film. Furthermore, the superfluid density decreases significantly with a corresponding decrease in film thickness. A 50 Å lead film was then grown, but this film was discontinuous and showed no superconducting transition. The percolation threshold must then occur at a film thickness between 50 and 100 Å, but there was not enough time to find the critical thickness.

As stated previously, $\lambda_{\perp}^{-1}(T)$ is expected to rise from zero proportional to $(1-T/T_c)^x$ at T_c for a system with a continuous phase transition. In order to determine the value of the exponent x, $\lambda_{\perp}^{-1}$ was divided by its derivative with respect to temperature, and the resultant value was plotted versus temperature for the three lead films. These graphs should give a line with slope 1/x (Figure 10).

Figure 10: Determination of exponent of $\lambda_{\perp}^{-1}$ for the phase transition
$\lambda_{\perp}^{-1}$ = two-dimensional magnetic penetration depth



The 100 Å and 150 Å graphs were linear within a range of about 0.05 K near their respective critical temperatures while the 575 Å film was linear within a range of about 0.03 K. The slopes of the three lines were 1.60 +/- 0.05 for the 100 Å film, 1.59 +/- 0.05 for the 150 Å film, and 1.20 +/- 0.05 for the 575 Å film. From the slopes for the thinner films, the change in $\lambda_\perp^{-1}$ is estimated to be proportional to $(1\text{-}T/T_c)^{5/8}$ or $(1\text{-}T/T_c)^{2/3}$. From the slope for the bulk lead, the change in $\lambda_\perp^{-1}$ is estimated to be proportional to $(1\text{-}T/T_c)^{5/6}$; however, mean field behavior in the bulk lead should cause the exponent to be 1. The difference between the theoretical and experimental values of the exponent indicates an error for which there is no explanation. Yet it can still be concluded that the thinner films differ even more from mean field behavior, since there is a larger temperature range of linearity in the $\lambda_\perp^{-1}$ / $d\{\lambda_\perp^{-1}(T)\}$ versus temperature graph for the thin films than for the thick film. Also, $\lambda_\perp^{-1}$ is proportional to a smaller exponent in the thin films than in the bulk lead film.

Conclusion:

A percolating lead system is difficult to study because of the possibility of lead migration over the surface of the film and because of the short thickness interval over which the film changes from completely discontinuous to completely continuous. In order to decrease the potential for lead migration, silicon was evaporated after lead in order to encapsulate it, but further study needs to be done to determine if silicon evaporation heat could actually induce lead migration, even if the substrate is being cooled by liquid nitrogen.

The thickness at which the percolation threshold occurs in lead films on silicon substrates has been narrowed to between 50 Å and 100 Å. A 50 Å film was found to be completely discontinuous, showing no superconducting phase transition. The 100 Å and 150 Å films had phase transitions over larger temperature intervals than the bulk lead film (575 Å), but it is expected that between 50 Å and 100 Å the phase transition will occur over a maximum temperature interval. The lead thickness at which this occurs is the percolation threshold. This hypothesis still remains to be tested.

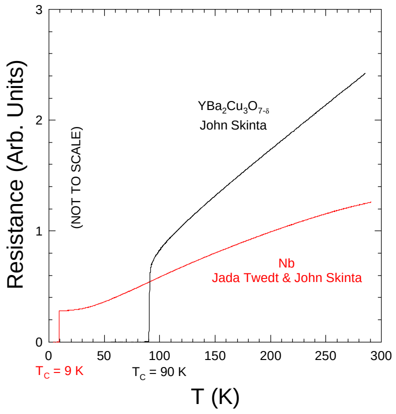
Finally, there appears to be no frequency dependence in the two-dimensional magnetic penetration depths for the continuous lead films, but frequency dependence should still be searched for in percolating systems. $\lambda_{\perp}^{-1}$ is found to rise from zero proportional to $(1\text{-}T/T_c)^{5/8}$ or $(1\text{-}T/T_c)^{2/3}$ for the thinner films and proportional to $(1\text{-}T/T_c)^{5/6}$ for the thick film as the temperature decreases through the transition. Measurements of other films with these same thicknesses must be taken in order to support these exponent values adequately. Also, the exponents for even thinner films must be measured to determine its limiting value at the percolation threshold.

Appendix A : The First Project

The proximity effect is the effect a magnetic or nonmagnetic layer has on the critical temperature and penetration depth of a superconductor. This additional layer may or may not be a superconductor itself. The proximity effect is important to understand for the practical applications of superconductors, since it is necessary to know the effects another metal has on a superconductor if the superconductor is to be used in electronics.

Originally, the proximity effect was to be studied by evaporating aluminum onto thin niobium films. The niobium films were produced by sputtering onto a silicon substrate, and the quality of the niobium was determined by the critical temperature (should be approximately 9 K) and the residual resistivity ratio (RRR). A pure metal will have zero resistance at 0 K. However, since this is impossible to measure, the ratio of the resistance at 300 K to that at 10 K is used as a measure of purity. A high quality niobium film has an RRR around 5. Figure 1 shows RRR measurements for both a YBCO film and a high quality niobium film. The critical temperature of the niobium is 9 K and the RRR is approximately 4.4. However, numerous problems with the sputtering system occurred, and the niobium film in Figure 11 was the only high quality film produced.

Figure 11: YBCO and Niobium Resistance Measurements



4 K : Helium Liquifies, LHe ~ $ 2 / Liter

77 K : Nitrogen Liquifies, LN_2 cheaper than Coke!

Appendix B: The Second Project

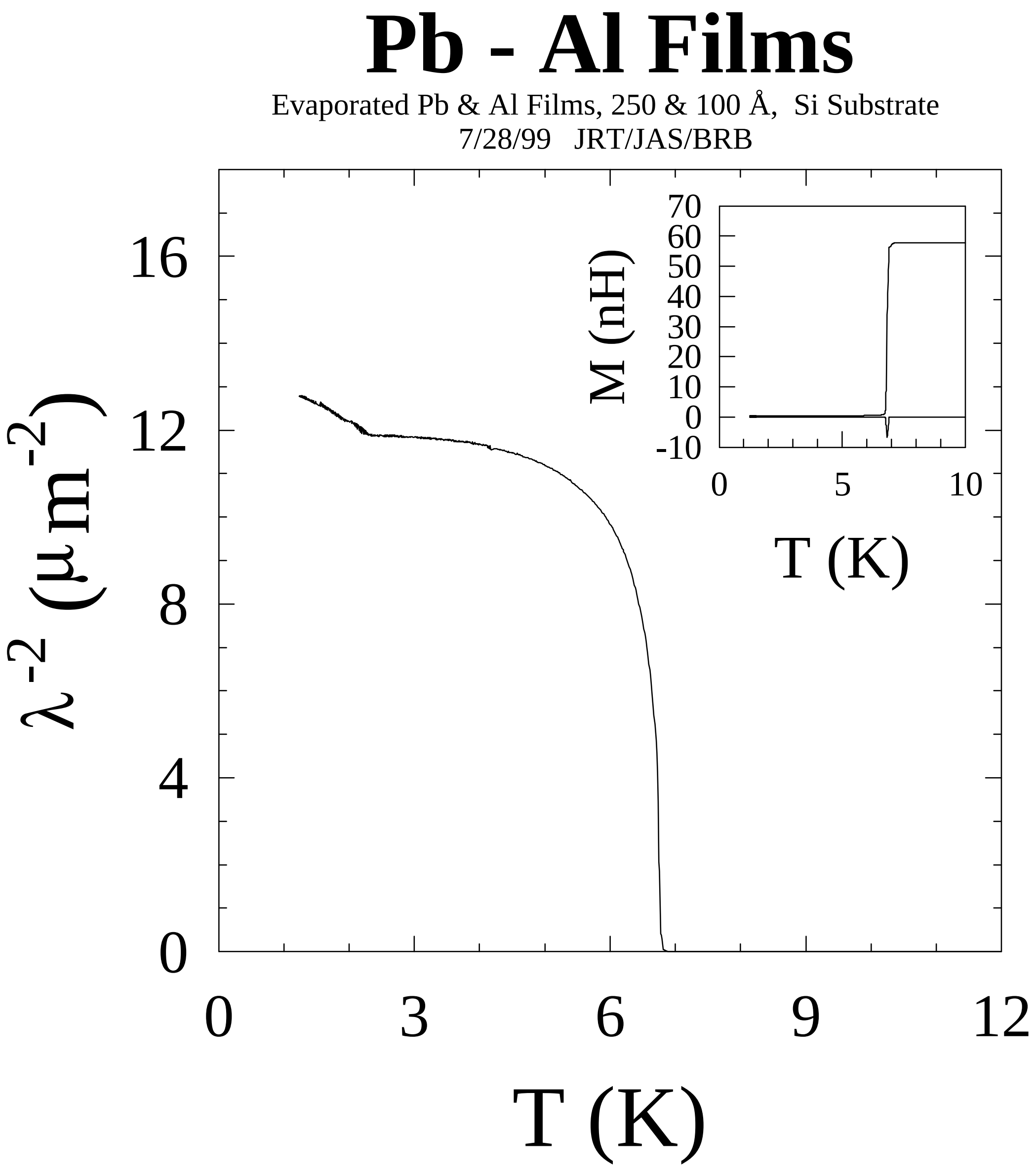
In order to create better films for studying the proximity effect, the coevaporator system was utilized to evaporate 100 Å of aluminum and then 250 Å of lead onto a silicon substrate. After the film was grown, a penetration depth was taken, and the plan was to grow other films with various thicknesses of aluminum and lead in order to compare the effect one metal has on the other metal's penetration depth and critical temperature.

Results of Second Project:

For this particular film, the transition for aluminum occurred at approximately 2 K, and the transition for lead occurred at approximately 6.7 K (Figure 12). At the transition for lead, the superfluid density does not increase smoothly as it should. Instead, there is a foot on the graph where the superfluid density appears to increase at a different rate as the temperature falls between 6.8 and 6.7 K. This behavior possibly corresponds to lead acting as a percolating system.

It is interesting that the percolation threshold for lead is between 50 and 100 Å on a bare silicon substrate, but by putting down 100 Å of aluminum first, percolation was possibly observed at a lead thickness of 250 Å. The effect of aluminum and other metals on percolating lead systems needs to be studied further.

Figure 12: Aluminum and Lead Film



Pb - Al Films

Evaporated Pb & Al Films, 250 & 100 Å, Si Substrate

7/28/99 JRT/JAS/BRB

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The expertise of Dr. Tom Lemberger, Brent Boyce, and John Skinta

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