Within the last funding period under AFOSR-90-0111, we made major discoveries in two areas. In superconducting thin films, the long range proximity effect and the observation of a novel resistance anomaly are new physical phenomena exposed through the development of a technique to precisely modify the transition temperature in a well-defined region. Free standing metallic structures were fabricated to understand thermal transport in thin films. In this latter work, the electron-phonon interaction should be modified due to the thermal cut-off of excitations. Using two different techniques, we have determined that the electron-phonon interaction does not display the expected thermal cut-off, a fact which may be of significance in thermalizing electronic components. We have also observed a reduction of the electron-impurity-spin interaction due to size effects, which should have interesting consequences for the magnetism of ultrasmall structures.
Terminal Report
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"Physics and Technology for the Investigation of Properties of Ultra Small Systems"

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ABSTRACT

Within the last funding period under AFOSR-90-0111, we made major discoveries in two areas. In superconducting thin films, the long range proximity effect and the observation of a novel resistance anomaly are new physical phenomena exposed through the development of a technique to precisely modify the transition temperature in a well defined region. Free standing metallic structures were fabricated to understand thermal transport in thin films. In this latter work, the electron-phonon interaction should be modified due to the thermal cut-off of excitations. Using two different techniques, we have determined that the electron-phonon interaction does not display the expected thermal cut-off, a fact which may be of significance in thermalizing electronic components. We have also observed a reduction of the electron-impurity-spin interaction due to size effects, which should have interesting consequences for the magnetism of ultrasmall structures.
Terminal report for activity under AFOSR Grant # 90-0111

We detail the research achieved in the course of the past two years. This work was carried out in collaboration with Professor Michael Isaacson and his students who have developed and made accessible much of the fabrication and characterization technology described in the text.

A. Anomalous Proximity Effect in Modulated T. Thin Film Structures

We have examined the anomalously long range superconducting proximity effect in a 2D analog of a modulated $T_c$ superlattice.\(^1\,^2\,^3\) The transition temperature was modulated by exposing sections of a $\sim 200\,\text{Å}$ thick aluminum film to a reactive ion-etch. The etch has the effect of modifying the transition temperature of the aluminum film by a few percent.\(^4\,^5\) The resulting structure has virtually identical normal state properties along its length; however, the transition temperature of the sections exposed to the etch is depressed by 2-5% relative to the unetched section.

In such a "superlattice," the resistive transition's character depends on the size of the normal region. Here we use normal to denote a superconducting metal above its transition temperature. In a normal metal, the proximity effect extends a distance of order $K_{n}^{-1}$, similar to the normal state coherence length $\xi_N = (\hbar D/2\pi k_B T)^{\frac{1}{2}}$. $K_{n}^{-1}$ diverges logarithmically near the transition temperature. In our samples, even at $T/T_c = 1.0$, \textit{this length is on the order of 1 $\mu$m.} Thus the expected range of the proximity effect in these relatively dirty aluminum films is on the order of 1 $\mu$m.\(^6\)

Figure 1. Resistive transitions for samples with unity ratio of etched to unetched material. In both (a) and (b), $\Diamond$ are $\Box$ and the uniformly etched and unetched films. In (a) resistive transitions shown are: $\times$, $+$, $\Delta$, $\circ$ for periodicities 4, 10, 20 and 100 $\mu$m. In (b) $\times$, $+$, $\Delta$, $\circ$ correspond to periodicity of 20, 100, 200 and 400 $\mu$m.
When we examined the resistive transitions of modulated transition temperature films, we found that the transitions were homogeneous, even when the intervening normal section was as long as 50 \( \mu \text{m} \) in length. Even at 200 \( \mu \text{m} \) the transitions were not "discrete" (figure 1). Although there have been reports in the literature of experiments which have propagated supercurrents across many coherence lengths\(^7\), all of those results show exponentially decreased critical currents. An examination of the critical currents of our modulated T\(_c\) films reveals robust superconductivity, comparable to that measured for the constituent sections.

The transition temperature of the modulated structures can be cast in a form suitable for superlattices\(^8\) where the modulation length scale is small compared to the coherence length (figure 2). The observation that our data fit this functional form together with the absence of the discrete transitions at length scales substantially longer than those expected from present theories led us to propose that the proximity effect extends over a length scale considerable longer than that previously observed, and that the mechanism of the proximity effect is not completely understood.

The question that arises, is why are our results so disparate from those of previous investigators who found excellent agreement with theory? The answer is undoubtedly bound to the similarity of the transition temperatures, and to the existence of virtually identical attractive potentials in the adjoining normal and superconducting materials. We have modelled the transition temperature of the system. These results also can be forced to give the appropriate transition temperatures only if the length scale over which pairing extends is adjusted to a value 20 times greater than that expected under the present theory.\(^6\)

![Figure 2. Reduced data for sets \( \circ, \square, \text{ and } \times \) with \( \Delta T_c = 23.8, 23.8 \) and 36.0 mK respectively. The lines are fit to the form \( \Delta T_c/(T_c-T_{c1}) = 1+pd_1/d_2 \) where 1 refers to the etched length and 2 to the unetched length. The values of \( p \) were 0.75, 0.9 and 1.7 respectively. The sets \( \circ, \) and \( \square \) had different periodicity.](image)

3
We patterned the films so that the etched and unetched sections ran parallel to the current path. If the proximity effect was due to non-equilibrium phenomena, then we would expect that the unetched sections would shunt the etched sections resulting in a constant, or nearly constant, higher $T_c$ film. To the contrary, we found that the transition temperature was virtually identical to that of films modulated in the perpendicular pattern in the same ratio. Thus, the transition temperature and superconductivity correspond to a thermodynamic transition.

We wished to investigate whether the periodic nature was responsible for the long length scale. Consequently, the film was patterned in the fibonacci sequence. We found that the resulting transition temperature had the same width and was located at the same transition temperature as a film with the same ratio of etched to unetched material.

We also have carried out preliminary investigations into the mechanisms responsible for the $T_c$ shift. By using micro-characterization techniques, (primarily Auger Electron Spectroscopy) we were able to establish that the main difference between the etched and the unetched section is that the oxygen present in the surface layer of the unetched film is replaced by fluorine (presumably from the breakdown of the freon etch). This substitution would lower $T_c$. The composition modulation closely follows the pattern that we lithographically defined, to within the spot size of the micro-characterization tool (2 $\mu$m). (figure 3)

![Figure 3. Line scan tracing the fluorine (a) and oxygen (b) Auger peak signals across several periods of etched and unetched aluminum film sections. The resolution is limited by the electron probe size.](image-url)
To summarize, this investigation has demonstrated that the long range proximity effect is present in materials with closely spaced transition temperatures and whose normal state properties are nearly identical. Clearly, the mechanisms for the maintenance of electron coherence in similar materials are not well understood. We note that there is at least one demonstration of a similar effect in composition modulated oxide superconductors. Thus there is overlap between our modulated $T_c$ film results and the oxide superconductors which are technologically important materials.

B. Resistive Anomaly at Normal-Superconducting Interfaces:

In this investigation, we sought to examine the potentials near a superconducting-normal interface, to determine if the spatial variation of these was responsible for the long range proximity effect described in the previous section. Aluminum was chosen as the metal to study since we had perfected the $T_c$ shift technique. We fabricated voltage leads symmetrically about the etched section. The transitions showed an anomalous increase in resistance prior to the drop at the superconducting transition temperature. (figure 4)

Figure 4. (a) The resistive transitions measured using voltage probes A through E (see inset) are shown as solid lines. The resistive anomaly is resolved by voltage leads A and B. In (b), we plot the normalized resistances shown in (a), for pairs A($\Theta$), B($\Delta$), C(+), D($\times$), and E($\circ$). The anomaly can be clearly observed at voltage leads A and B, and is absent for the outer leads. The magnitude of the anomaly decreases in both absolute and relative size with increasing distance from the N-S interface. In the inset of (a), a schematic diagram of the sample is shown. The central 50 $\mu$m section of the film is etched, depressing its $T_c$ by $\sim 45$ mK. Voltage lead pairs are located symmetrically about the etched-unetched interfaces and (in this case) are unetched. Pairs A, B, C, D, and E are positioned so that their inner edges are 0, 4, 9, 16 and 50 $\mu$m from the interface.
We have constructed a model which explains the main features of this phenomenon. Briefly, the necessary ingredients are a relatively long charge imbalance length, the length scale necessary for the quasiparticles injected into a superconductor from a normal section to relax into the equilibrium paired state. This length scale is on the order of a few microns in aluminum. If the normal state resistivity is low, then there exists a finite length near the S-N interface where the superconducting potential is greater than the corresponding normal state potential. Providing the measuring voltage leads are in the superconducting state, they will sample the superconducting potential, which can exceed the potential across the leads when the entire sample is in the normal state. Hence the anomalous resistance rise. We have never observed this resistance rise in structures where the voltage leads are etched (normal) which is entirely consistent with our model. This interesting interfacial phenomenon has been observed by two other groups one at IBM and the other in Germany. However, we are the first to provide an explanation for the effect and can eliminate it under appropriate circumstances. The proposed work concentrates on a more sophisticated version of this experiment and may have implications for the properties of normal metal-high T_c contacts. There is at least one similar observation in granular high T_c superconductors, though the anomalies probably originate from the inhomogeneous nature of the material.

C. The Electron-Phonon Interaction in Free Standing Films

In a sample of sufficiently small dimensions, the phonon spectrum which, in a bulk sample may be assumed as being continuous, has discrete energy states spaced linearly in energy. The spacing \( h\nu = hC_s k = hC_s 2\pi /d \) (where \( d \) is the sample thickness, \( C_s \) the speed of sound in the material) for a 100Å film is comparable to the thermal energy at a temperature near 10 K.

All previous investigations of the phonon spectrum were on samples mounted to substrates or rendered free standing by non-standard means. We have fabricated free standing samples with identical on substrate structures as controls. The effect of the substrate is ubiquitous. Since both the metal and substrate are solids, they have very similar sound velocities and the coupling across the metal-substrate interface is good. Consequently, it is reasonable to argue that the phonon spectrum in a supported metal film should not be materially different from that of a bulk sample. In an effort to manifest the expected thermal phonon cut-off, we decided to undertake two parallel investigations.
i) Measurement of the Inelastic Scattering rate in Free Standing Aluminum Films:

We constructed film having a 5 μm width and a 220Å thickness with a 50 μm length of substrate removed. Voltage lead pairs allowed us to measure the magnetoresistance of free standing and supported samples independently of one another. By fitting the magnetoresistance, we were able to determine the inelastic scattering rate at various temperatures up to 7.5 K. No a-priori constraints were placed on the scattering rates from free standing and supported samples at the same temperature. The results of the fits are shown in figure 5. There is no significant difference between the results of the free standing and supported samples. The inelastic scattering rate for both samples displays the $AT^3 + BT$ characteristic of the electron-phonon and electron-electron interactions in metals.

![Graph of inelastic scattering rate vs temperature](image)

Figure 5. The inelastic scattering rate for free standing (□) and supported (○) 22 nm thick aluminum films. The scattering rates are identical except for the difference induced near $T_c$.

The result is truly confounding. Here we have a system in which the phonon spectrum must reflect confinement effects. Contrary to the results obtained on substrates, there is no mechanism by which the phonon spectrum can be three dimensional. Yet, the characteristics of supported and free standing films display no differences other than a small shift in the transition temperature presumably due to strain effects. We conclude that the excitation of phonons in thin films is incompletely understood or, that there must exist other modes, which develop to compensate for finite size effects. Such surface modes must dominate at low temperatures. These measurements are carried out in equilibrium, ie. the temperature of the electrons and the phonons are nominally identical. Consequently, there is no escaping the conclusion that the electron-phonon interaction in the finite size domain is not well understood.
ii) The Electron-Phonon Interaction and Thermal Transport:

Thermalizing small devices is a continual interest to the electronics industry. The work that we have carried out has some bearing on this problem, and basically implies that small devices can rely on efficient thermal transport through the phonons irrespective of the size of the sample.

We have measured the thermal resistance between the electrons in our sample and the environment. Heat is generated in the electron gas due to inelastic scattering events that alter the energy that the electrons have acquired from the applied electric field. These "hot" electrons relax their energy into the phonon bath through electron-phonon scattering events. This scattering process is characterized by a thermal resistance. The phonons in turn transmit their excess energy to the substrate through the Kapitza boundary resistance. If there is no substrate, then the heat has to flow along the wire to the ends where thermalization must take place. The phonon conductivity down the wire may be taken to be negligible at temperatures below 10 K so the only mechanism for thermal transport down the wire is via electrons.

We had to develop an electron thermometer and elected to fabricate a dilute magnetic system whose resistance would show Kondo contributions at low temperatures (see next section). When heating the electrons, the shift in the resistance of the sample would provide us with a measure of the electron temperature. From the known power applied to the sample, and the temperature increase, we infer the thermal resistance of the electron gas to the substrate.

We found that for the supported sample the thermal resistance followed a $T^{-3}$ power law. When we measured the thermal resistance of the free standing film, we found that the behavior followed the $T^{-1}$ law expected for electron conductivity (figure 6). By immersing the sample inside helium fluid at a pressure greater than the critical pressure or by coating the samples with thin films of superfluid $^4$He, we restored the thermal path for the phonon channel in the free standing film. We observed that the free standing film's thermal resistance now coincided with that of the supported film at temperatures below 5 K. Also, the $T^{-3}$ power law was replicated for the free standing film in helium (figure 6).

The conclusion that we arrive at supports that from the magnetoresistance studies on the aluminum films. There appears to be no thermal cut-off of the phonon spectrum. Several additional approaches were tried. We have measured films having a film thickness of 1200Å. Such films should have a larger $k_F\ell$ and a longer mean free path. Also the
thermal cut-off should be shifted to lower temperatures. The thermal resistance that was measured was exactly in accord with that expected from the previous films. Finally, a study of small (1D) films whose characteristic width is (35nm) shows the same $T^{-3}$ behavior. Here we would have expected only 1D phonons to survive. A possible explanation is that new low energy surface modes compensate for the loss of low energy phonon modes and act as efficient electron scatterers.

Figure 6. The thermal resistance of 2000 ppm, 20nm thick x 2μm wide Cu:Cr films. (○) supported on substrate film shows $T^{-3}$ dependence, (△) free standing in vacuum shows $T^{-1}$ dependence. Free standing films (◇) 4He-film-coated and (□) in-bulk-4He, show identical thermal behavior to supported sample at low temperatures. Apparently the electron-phonon scattering process is not sensitive to phonon dimensionality.

These results are important in that they allow us to clearly separate the various contributions to thermalization of electrons in microfabricated structures. Particularly at moderately low temperatures, phonons are still a contributing mechanism to thermalization, and an understanding of the electron-phonon interaction is crucial for the heat removal from cold circuitry. From a physics standpoint, the electron-phonon interaction is very important, since it is the basis for superconductivity.

D. Suppression of the Kondo Effect and Magnetic Interactions by Finite Size

In the course of the study of the thermal resistance described above we had to select a material to use as an electron thermometer. We elected to use Cr in Cu as a suitable alloy primarily since it gave a large Kondo resistance for a relatively small impurity concentration. After fabricating films of appropriate thickness, we found that the magnitude of the Kondo resistance was far smaller than expected based on measurements of bulk systems (figure 7). Also, the resistance maximum associated with the spin glass transition was suppressed as a function of concentration and thickness.17
Since there have been questions raised in the literature\textsuperscript{18} about clustering of Cr atoms, we decided to pattern films of several widths in a single evaporation. We observed that the Kondo resistance did decrease with a characteristic width of approximately 1 \textmu m. Results on the thickness dependence by a group at Purdue University\textsuperscript{19} confirm our observation of the Kondo effect suppression in a quasi-1D geometry. Evidently, the effectiveness of the screening cloud that surrounds a magnetic impurity is altered when the host's dimension decreases. Small dilute magnetic systems are clearly more complicated when their dimensions are decreased down to the \textmu m size.

**Summary of Results For 90/91**

To sum up, the results from the past two years reflect the broad scope of the research that we have undertaken in the area of microfabricated systems. We have seen novel phenomena associated with the proximity effect and non-equilibrium behavior at S-N-S junctions. We fabricated free standing structures and explored the electron-phonon interaction and size effects in these samples. The development of an electron thermometer has exposed new size effects in the physics of magnetic interactions in small systems.
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