ELECTRON TUNNELING AND SUPERCONDUCTIVITY

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by

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In my laboratory notebook dated May 2, 1960 is the entry: "Friday, April 22, I performed the following experiment aimed at measuring the forbidden gap in a superconductor." This was obviously an extraordinary event not only because I rarely write in my notebook, but because the success of that experiment is the reason I have the great honor and pleasure of addressing you today. I shall try in this lecture, as best I can, to recollect some of the events and thoughts that led to this notebook entry, though it is difficult to describe what now appears to me as fortuitous. I hope that this personal and subjective recollection will be more interesting to you than a strictly technical lecture, particularly since there are now so many good review articles dealing with superconductive tunneling. [1,2]

A recent headline in an Oslo paper read approximately as follows: "Master in billiards and bridge, almost flunked physics - gets Nobel Prize." The paper refers to my student days in Trondheim. I have to admit that the reporting is reasonably accurate, therefore I shall not attempt a "cover up", but confess that I almost flunked in mathematics as well. In those days I was not very interested in mechanical engineering and school in general, but I did manage to graduate with an average degree in 1952. Mainly because of the housing shortage which existed in Norway, my wife and I finally decided to emigrate to Canada where I soon found employment with Canadian General Electric. A three year Company course in engineering and applied mathematics known as the A, B and C course was offered to me. I realized this time that school was for real, and since it probably would be my last chance, I really studied hard for a few years.

When I was 28 years old I found myself in Schenectady, New York where I discovered that it was possible for some people to make a good living as physicists. I had worked on various Company assignments in applied mathematics, and had developed the feeling that the mathematics was much more advanced than the actual knowledge of the physical systems that we applied it to. Thus, I thought perhaps I should learn some physics and, even though I was still an engineer, I was given the opportunity to try it at the General Electric Research Laboratory.

The assignment I was given was to work with thin films and to me films meant photography. However I was fortunate to be associated with John Fisher who obviously had other things in mind. Fisher had started out as a mechanical engineer as well, but had lately turned his atten-

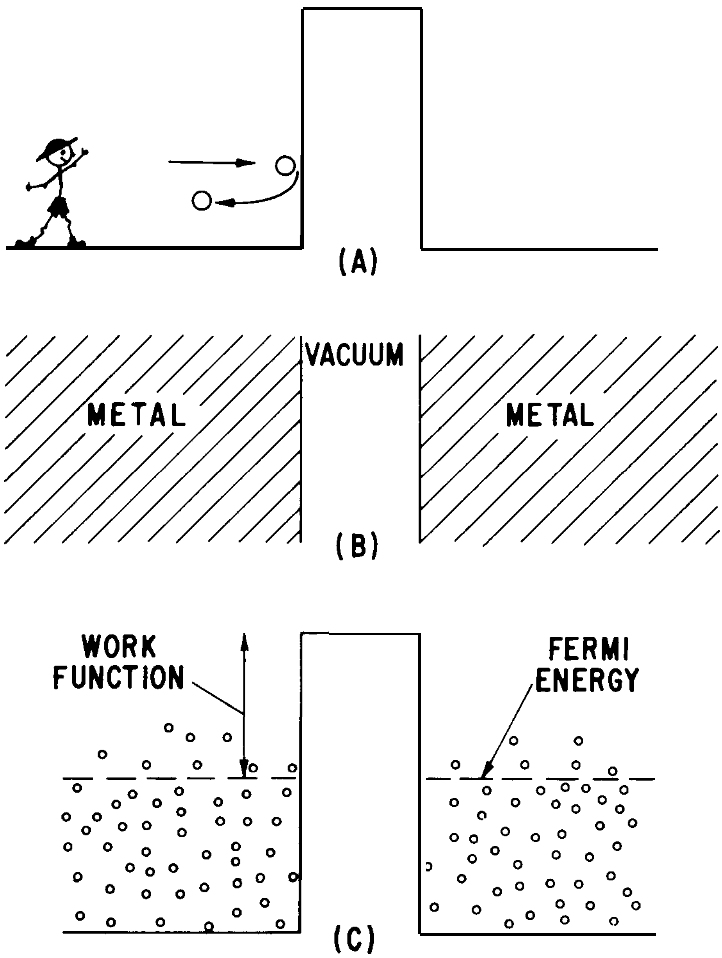

Fig. 1.
A. If a man throws a ball against a wall the ball bounces back. The laws of physics allow the ball to penetrate or tunnel through the wall but the chance is infinitesimally small because the ball is a macroscopic object. B. Two metals separated by a vacuum will approximate the above situation. The electrons in the metals are the "balls", the vacuum represents the wall. C. A pictorial energy diagram of the two metals. The electrons do not have enough energy to escape into the vacuum. The two metals can, however, exchange electrons by tunneling. If the metals are spaced close together the probability for tunneling is large because the electron is a microscopic particle.

tion towards theoretical physics. He had the notion that useful electronic devices could be made using thin film technology and before long I was working with metal films separated by thin insulating layers trying to do tunneling experiments. I have no doubt that Fisher knew about Leo Esaki's tunneling experiments at that time, but I certainly did not. The concept that a particle can go through a barrier seemed sort of strange to me, just struggling with quantum mechanics at Rensselaer Polytechnic Institute in Troy, where I took formal courses in Physics. For an engineer it sounds rather strange that if you

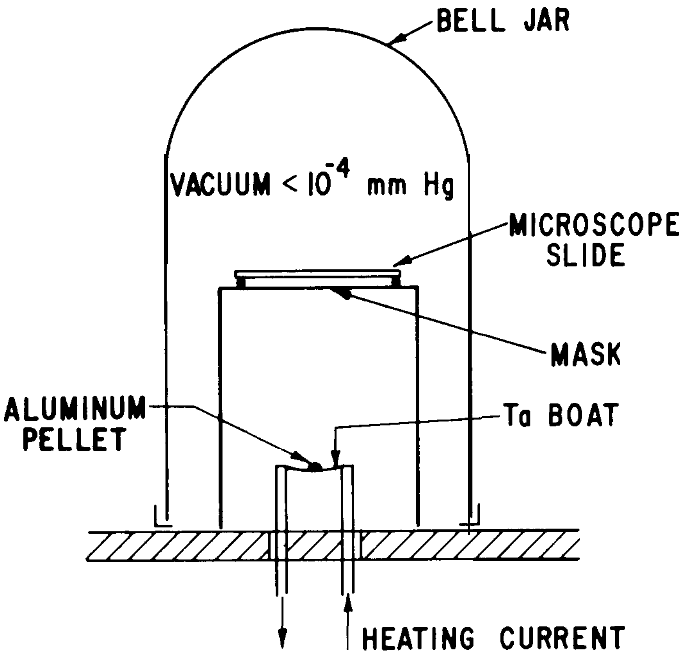

Fig. 2.
A schematic drawing of a vacuum system for depositing metal films. For example, if aluminum is heated resistively in a tantalum boat, the aluminum first melts, then boils and evaporates. The aluminum vapor will solidify on any cold substrate placed in the vapor stream. The most common substrates are ordinary microscope glass slides. Patterns can be formed on the slides by suitably shielding them with a metal mask.

throw a tennis ball against a wall enough times it will eventually go through without damaging either the wall or itself. That must be the hard way to a Nobel Prize! The trick, of course, is to use very tiny balls, and lots of them. Thus if we could place two metals very close together without making a short, the electrons in the metals can be considered as the balls and the wall is represented by the spacing between the metals. These concepts are shown in Figure 1. While classical mechanics correctly predicts the behavior of large objects such as tennis balls, to predict the behavior of small objects such as electrons we must use quantum mechanics. Physical insight relates to everyday experiences with large objects, thus we should not be too surprised that electrons sometimes behave in strange and unexpected ways.

Neither Fisher nor I had much background in experimental physics, none to be exact, and we made several false starts. To be able to measure a tunneling current the two metals must be spaced no more than about 100 Å apart, and we decided early in the game not to attempt to use air or vacuum between the two metals because of problems with vibration. After all, we both had training in mechanical engineering! We tried instead to keep the two metals apart by using a variety of thin insulators made from Langmuir films and from Formvar. Invariably, these films had pinholes and the mercury

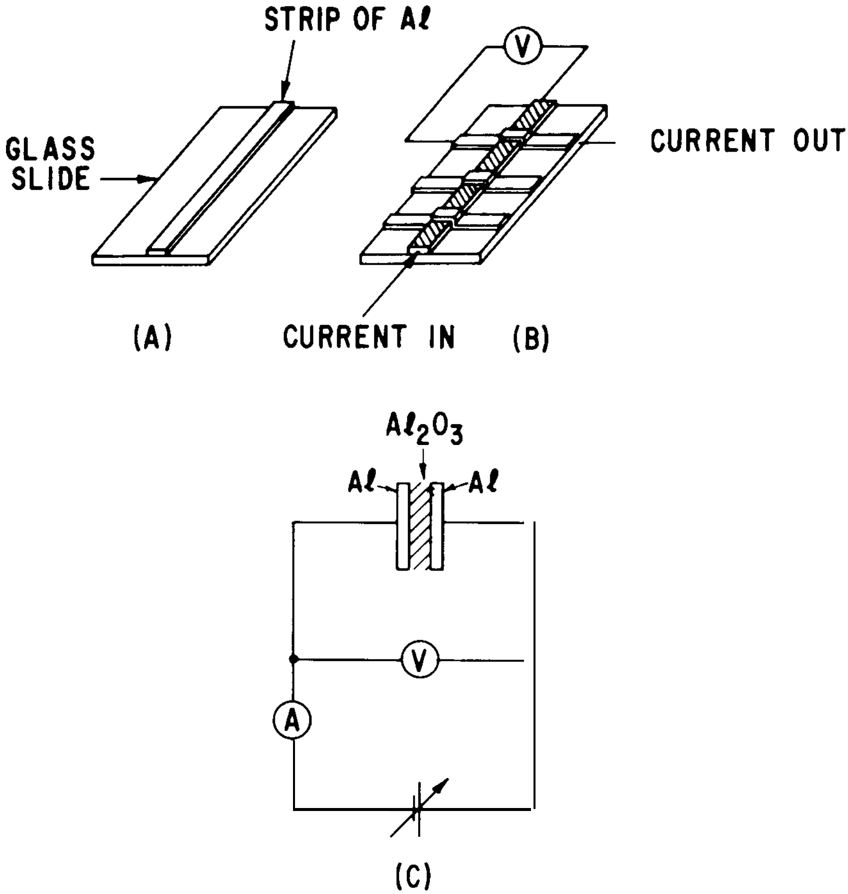

Fig. 3.
A. A microscope glass slide with a vapor deposited aluminum strip down the middle. As soon as the aluminum film is exposed to air, a protective insulating oxide forms on the surface. The thickness of the oxide depends upon such factors as time, temperature and humidity. B. After a suitable oxide has formed, cross strips of aluminum are evaporated over the first film, sandwiching the oxide between the two metal films. Current is passed along one aluminum film up through the oxide and out through the other film, while the voltage drop is monitored across the oxide. C. A schematic circuit diagram. We are measuring the current-voltage characteristics of the capacitor-like arrangement formed by the two aluminum films and the oxide. When the oxide thickness is less than 50Å or so, an appreciable dc current will flow through the oxide.

counter electrode which we used would short the films. Thus we spent some time measuring very interesting but always non-reproducible current-voltage characteristics which we referred to as miracles since each occurred only once. After a few months we hit on the correct idea: to use evaporated metal films and to separate them by a naturally grown oxide layer.

To carry out our ideas we needed an evaporator, thus I purchased my first piece of experimental equipment. While waiting for the evaporator to arrive I worried a lot-1 was afraid I would get stuck in experimental physics tied

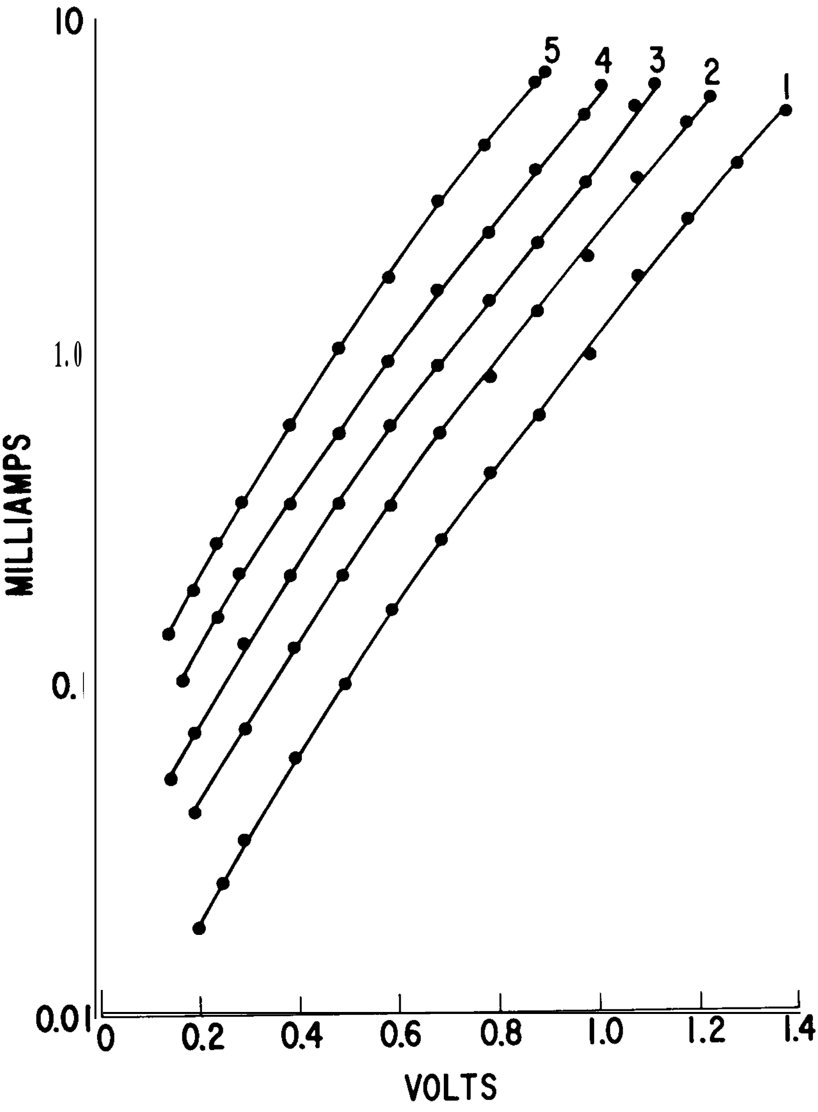

Fig. 4.
Current-voltage characteristics of five different tunnel junctions all with the same thickness, but with five different areas. The current is proportional to the area of the junction. This was one of the first clues that we were dealing with tunneling rather than shorts. In the early experiments we used a relatively thick oxide, thus very little current would flow at low voltages.

down to this expensive machine. My plans at the time were to switch into theory as soon as I had acquired enough knowledge. The premonition was correct; I did get stuck with the evaporator, not because it was expensive but because it fascinated me. Figure 2 shows a schematic diagram of an evaporator. To prepare a tunnel junction we first evaporated a strip of aluminum onto a glass slide. This film was removed from the vacuum system and heated

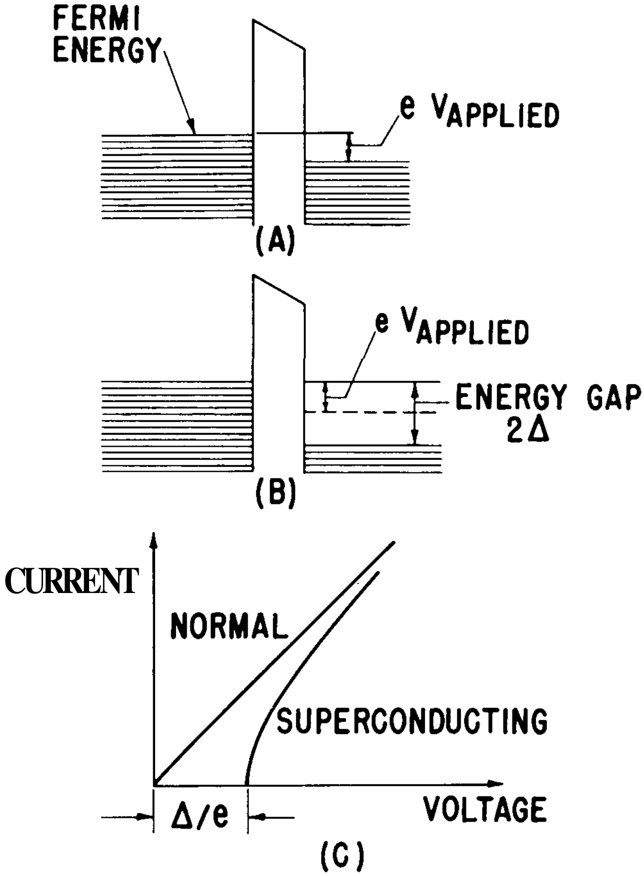

Fig. 5.
A. An energy diagram of two metals separated by a barrier. The Fermi energies in the two metals are at different levels because of the voltage difference applied between the metals. Only the left metal electrons in the energy range $e \, . \, V_{\mathrm{App}}$ can make a transition to the metal on the right, because only these electrons face empty energy states. The Pauli Principle allows only one electron in each quantum state. B. The right-hand metal is now superconducting, and an energy gap 2Δ has opened up in the electron spectrum. No single electron in a superconductor can have an energy such that it will appear inside the gap. The electrons from the metal on the left can still tunnel through the barrier, but they cannot enter into the metal on the right as long as the applied voltage is less than Δ/e, because the electrons either face a filled state or a forbidden energy range. When the applied voltage exceeds Δ/e, current will begin to flow. C. A schematic current-voltage characteristic. When both metals are in the normal state the current is simply proportional to the voltage. When one metal is superconducting the current-voltage characteristic is drastically altered. The exact shape of the curve depends on the electronic energy spectrum in the superconductor.

to oxidize the surface rapidly. Several cross strips of aluminum were then deposited over the first film making several junctions at the same time. The steps in the sample preparation are illustrated in Figure 3. This procedure solved two problems, first there were no pinholes in the oxide because it is

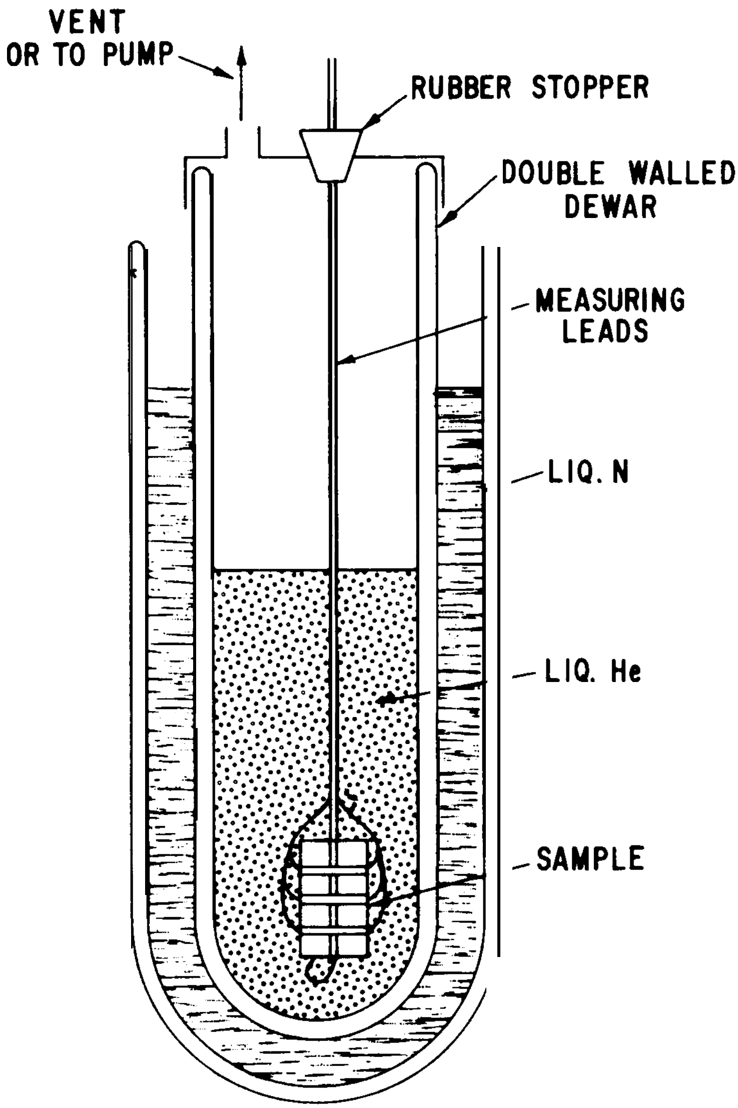

Fig. 6.
A standard experimental arrangement used for low temperature experiments. It consists of two dewars, the outer one contains liquid nitrogen, the inner one, liquid helium. Helium boils at 4.2" K at atmospheric pressure. The temperature can be lowered to about 1°K by reducing the pressure. The sample simply hangs into the liquid helium supported by the measuring leads.

self-healing, and second we got rid of mechanical problems that arose with the mercury counter electrode.

By about April, 1959, we had performed several successful tunneling experiments. The current-voltage characteristics of our samples were reasonably reproducible, and conformed well to theory. A typical result is shown in Figure 4. Several checks were done, such as varying the area and the oxide thickness of the junction as well as changing the temperature. Everything looked OK, and I even gave a seminar at the Laboratory. By this time, I had solved Schrodinger's equation enough times to believe that electrons sometimes behave as waves, and I did not worry much about that part anymore.

However: there were many real physicists at the Laboratory and they properly questioned my experiment. How did I know I did not have metallic shorts? Ionic current? Semiconduction rather than tunneling3 Of course, I did not know, and even though theory and experiments agreed well, doubts about the validity were always in my mind. I spent a lot of time inventing impossible schemes such as a tunnel triode or a cold cathode, both to try to prove conclusively that I dealt with tunneling and to perhaps make my work useful. It was rather strange for me at that time to get paid for doing what I considered having fun, and my conscience bothered me. But just like quantum mechanics, you get used to it, and now I often argue the opposite point; we should pay more people to do pure research.

I continued to try out my ideas on John Fisher who was now looking into the problems of fundamental particles with his characteristic optimism and enthusiasm; in addition, I received more and more advice and guidance from Charles Bean and Walter Harrison, both physicists with the uncanny ability of making things clear as long as a piece of chalk and a blackboard were available. I continued to take formal courses at RPI, and one day in a solid state physics course taught by Professor Huntington we got to superconductivity. Well, I didn't believe that the resistance drops to exactly zero-but what really caught my attention was the mention of the energy gap in a superconductor, central to the new Bardeen-Cooper-Schrieffer theory. If the theory was any good and if my tunneling experiments were any good, it was obvious to me that by combining the two, some pretty interesting things should happen, as illustrated in Figure 5. When I got back to the GE Laboratory I tried this simple idea out on my friends, and as I remember, it did not look as good to them. The energy gap was really a many body effect and could not be interpreted literally the way I had done. But even though there was considerable skepticism, everyone urged me to go ahead and make a try. Then I realized that I did not know what the size of the gap was in units I understood-electron volts. This was easily solved by my usual method: first asking Bean and then Harrison, and, when they agreed on a few millielectron volts: I was happy because that is in a easily measured voltage range.

I had never done an experiment requiring low temperatures and liquid helium-that seemed like complicated business. However one great advantage of being associated with a large laboratory like General Electric is that there are always people around who are knowledgeable in almost any field, and better still they are willing to lend you a hand. In my case, all I had to do was go to the end of the hall where Warren DeSorbo was already doing experiments with superconductors. I no longer remember how long it took me to set up the helium dewars I borrowed, but probably no longer than a day or two. People unfamiliar with low temperature work believe that the whole field of low temperature is pretty esoteric, but all it really requires is access

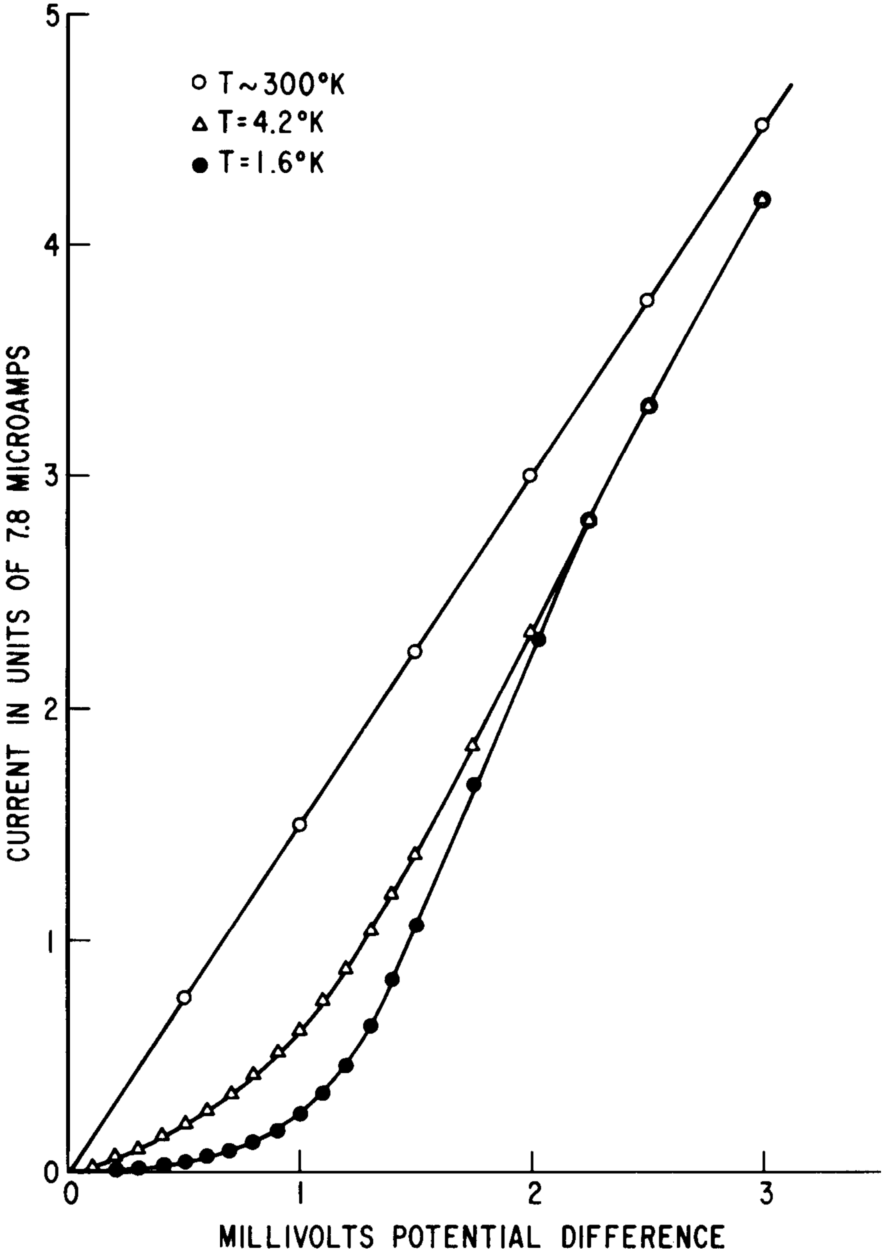

Fig. 7.
The current-voltage characteristic of an aluminum-aluminum oxide-lead sample. As soon as the lead becomes superconducting the current ceases to be proportional to the voltage. The large change between 4.2" K and 1.6" K is due to the change in the energy gap with temperature. Some current also flows at voltages less than Δ / e because of thermally excited electrons in the conductors.

to liquid helium, which was readily available at the Laboratory. The experimental setup is shown in Figure 6. Then I made my samples using the familiar aluminum-aluminum oxide, but I put lead strips on top. Both lead and alu-

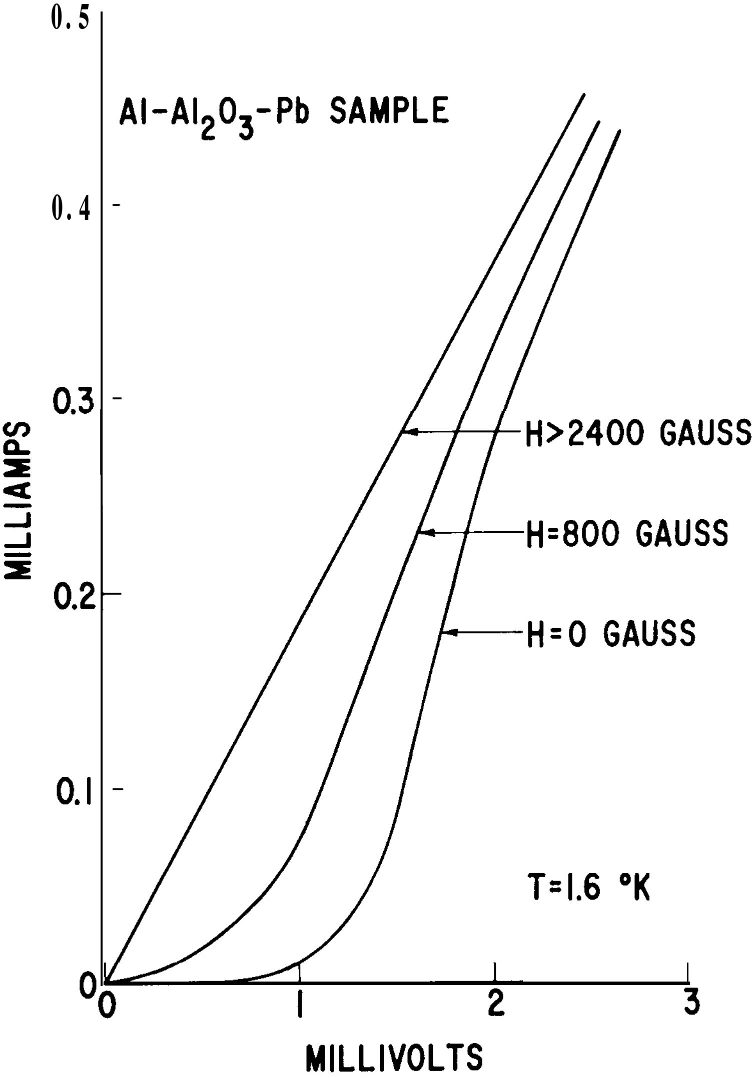

Fig. 8.
The current-voltage characteristic at 1.6" K as a function of the applied magnetic field. At 2 400 gauss the films are normal, at 0 gauss the lead film is superconducting. The reason for the change in the characteristics between 800 gauss and 0 gauss is that thin films have an energy gap that is a function of the magnetic field.

minum are superconductors, lead is superconducting at 7.2° K and thus all you need to make it superconducting is liquid helium which boils at 4.2° K. Aluminum becomes superconducting only below 1.2° K, and to reach this temperature a more complicated experimental setup is required.

The first two experiments I tried were failures because I used oxide layers which were too thick. I did not get enough current through the thick oxide to measure it reliably with the instruments I used, which were simply a standard voltmeter and a standard ammeter. It is strange to think about that

Fig. 9.
Informal discussion over a cup of coffee. From left: Ivar Giaever, Walter Harrison, Charles Bean, and John Fisher.

now, only 13 years later, when the Laboratory is full of sophisticated x-y recorders. Of course, we had plenty of oscilloscopes at that time but I was not very familiar with their use. In the third attempt rather than deliberately oxidizing the first aluminum strip, I simply exposed it to air for only a few minutes, and put it back in the evaporator to deposit the cross strips of lead. This way the oxide was no more than about 30Å thick, and I could readily measure the current-voltage characteristic with the available equipment. To me the greatest moment in an experiment is always just before I learn whether the particular idea is a good or a bad one. Thus even a failure is exciting, and most of my ideas have of course been wrong. But this time it worked! The current-voltage characteristic changed markedly when the lead changed from the normal state to the superconducting state as shown in Figure 7. That was exciting! I immediately repeated the experiment using a different sample - everything looked good! But how to make certain? It was well-known that superconductivity is destroyed by a magnetic field, but my simple setup of dewars made that experiment impossible. This time I had to go all the way across the hall where Israel Jacobs studied magnetism at low temperatures. Again I was lucky enough to go right into an experimental rig where both the temperature and the magnetic field could be controlled and I could quickly do all the proper experiments. The basic result is shown in Figure 8. Every-

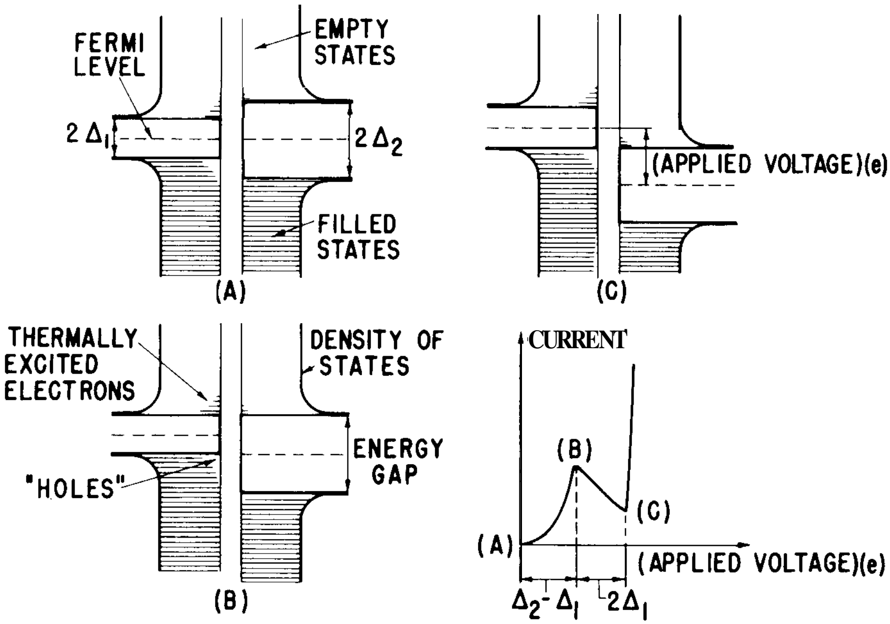

Fig. 10.
Tunneling between two superconductors with different energy gaps at a temperature larger than 0° K. A. No voltage is applied between the two conductors. B. As a voltage is applied it becomes energetically possible for more and more of the thermally excited electrons to flow from the superconductor with the smaller gap into the superconductor with the larger gap. At the voltage shown all the excited electrons can find empty states on the right. C. As the voltage is further increased, no more electrons come into play, and since the number of states the electrons can tunnel into decreases, the current will decrease as the voltage is increased. When the voltage is increased sufficiently the electrons below the gap in the superconductor on the left face empty states on the right, and a rapid increase in current will occur. D. A schematic picture of the expected current-voltage characteristic.

thing held together and the whole group, as I remember it, was very excited. In particular, I can remember Bean enthusiastically spreading the news up and down the halls in our Laboratory, and also patiently explaining to me the significance of the experiment.

I was, of course, not the first person to measure the energy gap in a superconductor, and I soon became aware of the nice experiments done by M. Tinkham and his students using infrared transmission. I can remember that I was worried that the size of the gap that I measured did not quite agree with those previous measurements. Bean set me straight with words to the effect that from then on other people would have to agree with me; my experiment would set the standard, and I felt pleased and like a physicist for the first time.

That was a very exciting time in my life; we had several great ideas to improve and extend the experiment to all sorts of materials like normal metals, magnetic materials and semiconductors. I remember many informal dis-

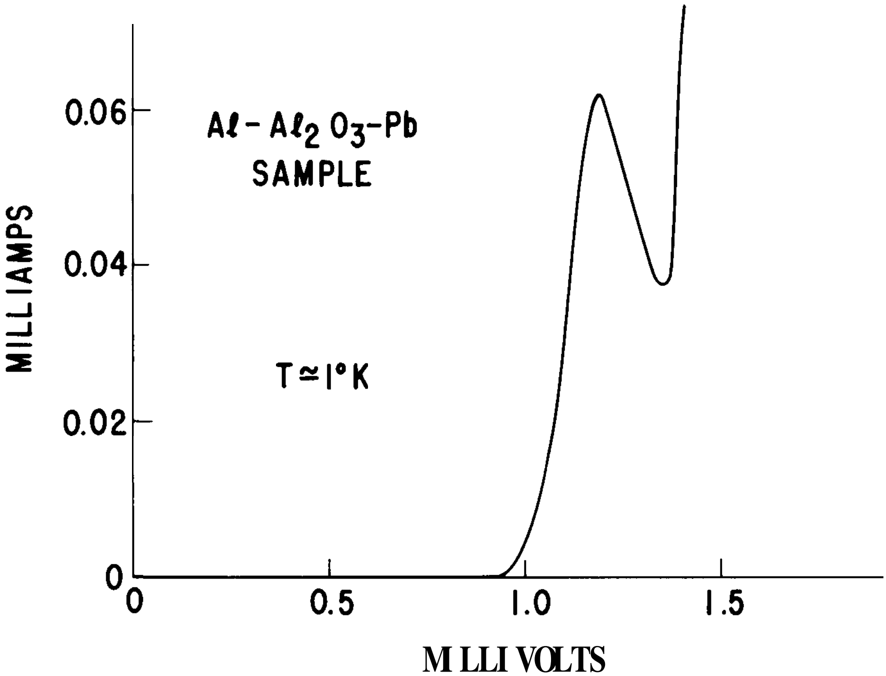

Fig. 11.
A negative resistance characteristic obtained experimentally in tunneling between two different superconductors.

cussions over coffee about what to try next and one of these sessions is in a photograph taken in 1960 which is shown in Figure 9. To be honest the picture was staged, we weren't normally so dressed up, and rarely did I find myself in charge at the blackboard! Most of the ideas we had did not work very well and Harrison soon published a theory showing that life is really complicated after all. But the superconducting experiment was charmed and always worked. It looked like the tunneling probability was directly proportional to the density of states in a superconductor. Now if this were strictly true, it did not take much imagination to realize that tunneling between two superconductors should display a negative resistance characteristic as illustrated in Figure 10. A negative resistance characteristic meant, of course, amplifiers, oscillators and other devices. But nobody around me had facilities to pump on the helium sufficiently to make aluminum become superconducting. This time I had to leave the building and reactivate an old low temperature setup in an adjacent building. Sure enough, as soon as the aluminum went superconducting a negative resistance appeared, and, indeed, the notion that the tunneling probability was directly proportional to the density of states was experimentally correct. A typical characteristic is shown in Figure 11.

Now things looked very good because all sorts of electronic devices could be made using this effect, but, of course, they would only be operative at low temperatures. We should remember that the semiconducting devices were not so advanced in 1960 and we thought that the superconducting junction

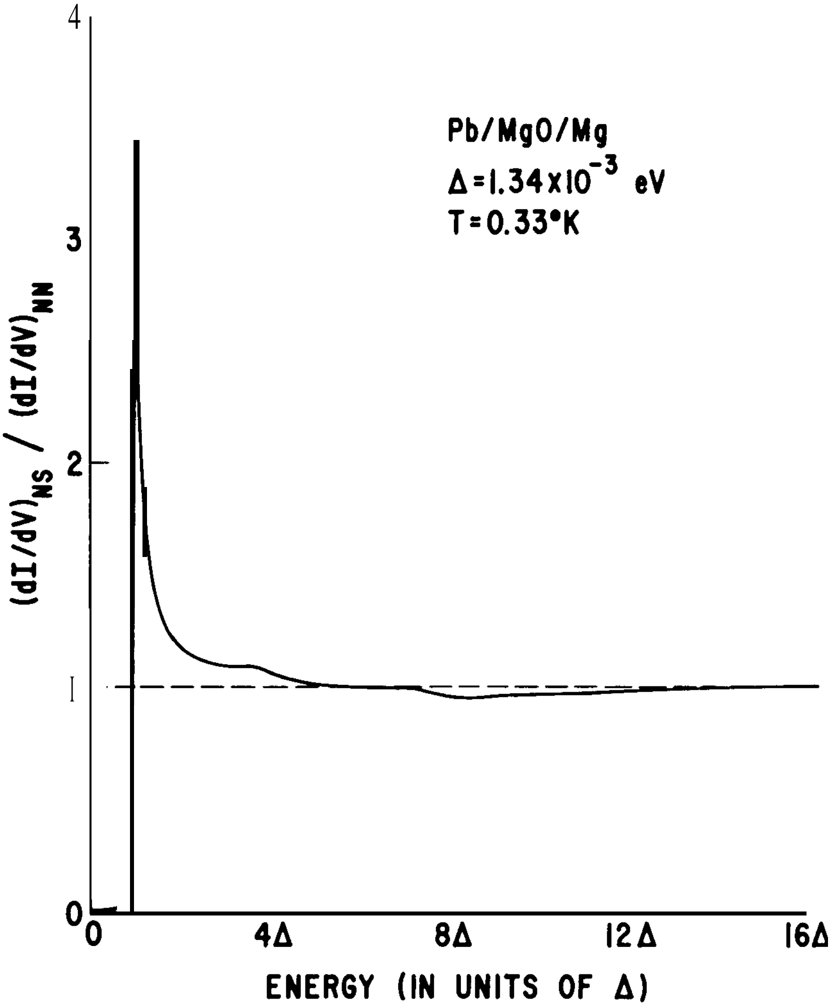

Fig. 12.
A normalized derivative of the current with respect to voltage of a lead junction at low temperature. The simple BCS-theory predicts that the derivative should approach unity asymptotically as the energy increases. Instead several wiggles are observed in the range between 4Δ and 8Δ. These wiggles are related to the phonon spectrum in lead.

would have a good chance of competing with, for example, the Esaki diode. The basic question I faced was which way to go: engineering or science? I decided that I should do the science first, and received full support from my immediate manager, Roland Schmitt.

In retrospect I realize how tempting it must have been for Schmitt to encourage other people to work in the new area, and for the much more experienced physicists around me to do so as well. Instead, at the right time, Schmitt provided me with a co-worker, Karl Megerle, who joined our Laboratory as a Research Training Fellow. Megerle and I worked well together and before long we published a paper dealing with most of the basic effects.

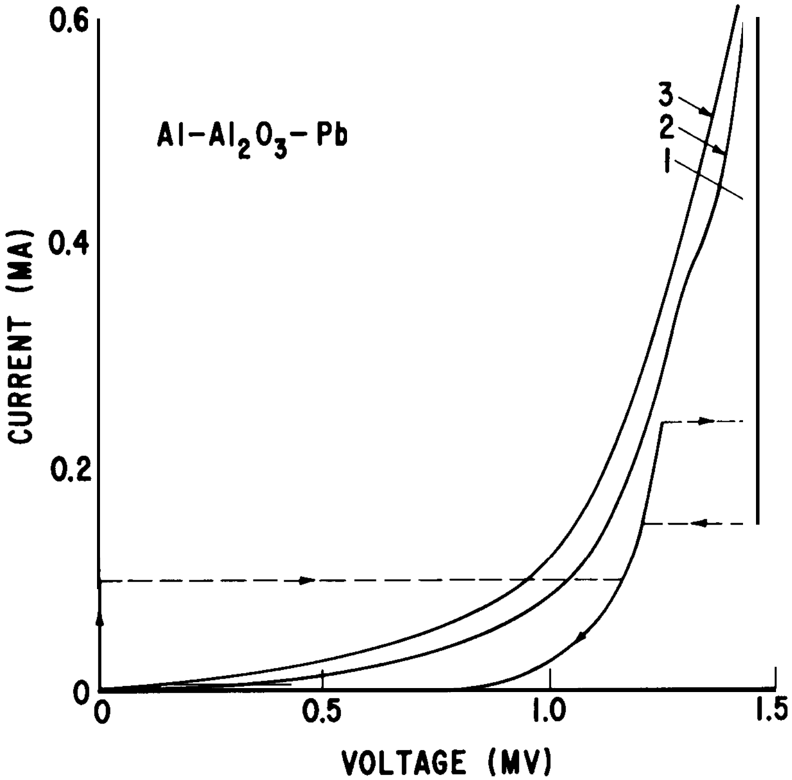

Fig. 13.
Effect of trapped magnetic field on a tunneling characteristic. Curve 1 is a virgin curve, while curve 3 is in a moderate magnetic field, and in curve 2 the magnetic field has been removed. In curve 1 we also have a small resistance-less current which we interpreted as caused by metallic shorts. In retrospect, it was actually due to the Josephson effect.

As always in physics, it is important to extend experiments to a higher energy, a greater magnetic field, or, in our case, to a lower temperature. Therefore, we joined forces with Howard Hart, who had just completed a helium 3 refrigerator that was capable of getting down to about 0.3° K. At the same time, Megerle finished a lock-in amplifier which we could use to measure directly the derivative of the current with respect to the voltage. That was really a nice looking machine with a magnet rotating past a pickup coil at eight cycles per second, but, of course, vastly inferior to the modern lock-in amplifier. We had known for some time that there were anomalies in the current-voltage characteristics of lead, and now we finally pinned them down by finding some extra wiggles in the derivative curve. This is shown in Figure 12. That made us happy because all that the tunneling experiments had done up till now was to confirm the BCS theory, and that is not what an experimentalist would really like to do. The dream is to show that a famous theory is incorrect, and now we had finally poked a hole in the theory. We speculated at the time that these wiggles were somehow associated with the phonons thought to be the cause of the attractive electron-electron interaction in a superconductor. As often happens, the theorists turned the tables on us and cleverly used these wiggles to properly extend the theory and to prove that the BCS theory indeed was correct. Professor Bardeen gave a detailed account of this in his most recent Nobel Prize lecture.

I have, so far, talked mainly about what went on at General Electric at that time; sometimes it is difficult for me to realize that Schenectady is not the center of the world. Several other people began to do tunneling work, and to mention just a few: J. M. Rowell and W. L. McMillan were really the ones who unraveled the phonon structure in a superconductor; W. J. To - masch, of course, insisted on discovering his own effect; S. Shapiro and colleagues did tunneling between two superconductors at the same time we did; and J. Bardeen, and later M. H. Cohen et al., took care of most of the theory.

Meanwhile, back at RPI, I had finished my course work and decided to do a theoretical thesis on ordered-disordered alloys with Professor Huntington because tunneling in superconductors was mainly understood. Then someone made me aware of a short paper by Brian Josephson in *Physics Letters* - what did I think? Well, I did not understand the paper, but shortly after I had the chance to meet Josephson at Cambridge and I came away impressed. One of the effects Josephson predicted was that it should be possible to pass a supercurrent with zero voltage drop through the oxide barrier when the metals on both sides were superconducting; this is now called the dc Josephson effect. We had observed this behavior many times; matter-of-fact, it is difficult not to see this current when junctions are made of tin-tin oxide-tin or lead-lead oxide-lead. The early tunnel junctions were usually made with aluminum oxide which generally is thicker and therefore thermal fluctuations suppress the dc current. In our first paper Megerle and I published a curve, which is shown in Figure 13, demonstrating such a supercurrent and also that it depended strongly on a magnetic field. However, I had a ready-made explanation for this supercurrent-it came from a metallic short or bridge. I was puzzled at the time because of the sensitivity to the magnetic field which is unexpected for a small bridge, but no one knew how a 20Å long and 20Å, wide bridge would behave anyway. If I have learned anything as a scientist it is that one should not make things complicated when a simple explanation will do. Thus all the samples we made showing the Josephson effect were discarded as having shorts. This time I was too simple-minded! Later I have been asked many times if I feel bad for missing the effect? The answer is clearly no, because to make an experimental discovery it is not enough to observe something, one must also realize the significance of the observation, and in this instance I was not even close. Even after I learned about the dc Josephson effect, I felt that it could not be distinguished from real shorts, therefore I erroneously believed that only the observation of the so-called ac effect would prove or disprove Josephson's theory.

In conclusion I hope that this rather personal account may provide some slight insight into the nature of scientific discovery. My own beliefs are that the road to a scientific discovery is seldom direct, and that it does not necessarily require great expertise. In fact, I am convinced that often a newcomer to a field has a great advantage because he is ignorant and does not know all the complicated reasons why a particular experiment should not be attempted. However, it is essential to be able to get advice and help from experts in the various sciences when you need it. For me the most important ingredients were that I was at the right place at the right time and that I found so many friends both inside and outside General Electric who unselfishly supported me.

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ENERGY GAP IN SUPERCONDUCTORS MEASURED BY ELECTRON TUNNELING

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If a potential difference is applied to two metals separated by a thin insulating film, a current will flow because of the ability of electrons to penetrate a potential barrier. The fact that for low fields the tunneling current is proportional to the applied voltage[1] suggested that low-voltage tunneling experiments could reveal something of the electronic structure of superconductors.

Aluminum/aluminum oxide/lead sandwiches were prepared by vapor-depositing aluminum on glass slides in vacuum, oxidizing the aluminum in air for a few minutes at room temperature, and then vapor-depositing lead over the aluminum oxide. The oxide layer separating aluminum and lead is thought to be about 15-20A thick.

At liquid helium temperature, in the presence of a magnetic field applied parallel to the film and sufficiently strong to keep the lead in the normal state, the tunnel current is linear in the voltage. However, when the magnetic field is removed, and lead becomes superconducting, the tunnel current is very much reduced at low voltages as shown in Fig. 1. There is no influence of polarity, identical results being obtained with both directions of current flow.

The slope dI/dV of the curve in Fig. 1 where $H=0$, $T=1.6°K$, divided by dI/dV for normal lead, is plotted in Fig. 2. On the naive picture that tunneling is proportional to density of states,[2] this curve expresses the density of states in superconducting lead relative to the density of states when lead is in its normal state, as a function of energy measured from the Fermi energy. It seems clear that the density of states at the Fermi level is drastically changed when a metal becomes a superconductor, the change being symmetric with respect to the Fermi level. The curve resembles the Bardeen-Cooper-Schrieffer[3] density of states for quasi-particle excitations. There is a broadening of the peak that decreases with decreasing temperature.

An approximate measure of half the energy gap

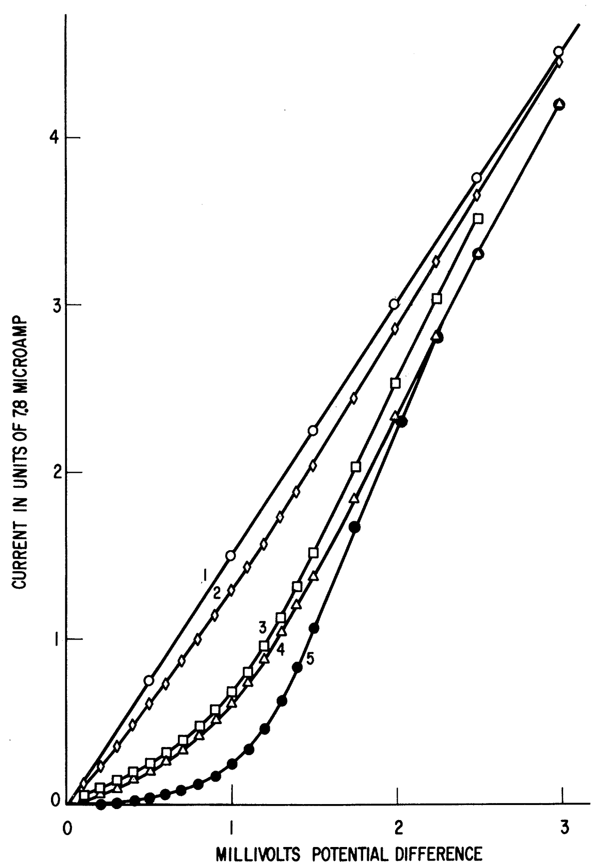

FIG. 1. Tunnel current between Al and Pb through Al_2O_3 film as a function of voltage. (1) $T=4.2°K$ and $1.6°K$, $H=2.7$ koe (Pb normal). (2) $T=4.2°K$, $H=0.8$ koe. (3) $T=1.6°K$, $H=0.8$ koe. (4) $T=4.2°K$, $H=0$ (Pb superconducting). (5) $T=1.6°K$, $H=0$ (Pb superconducting).

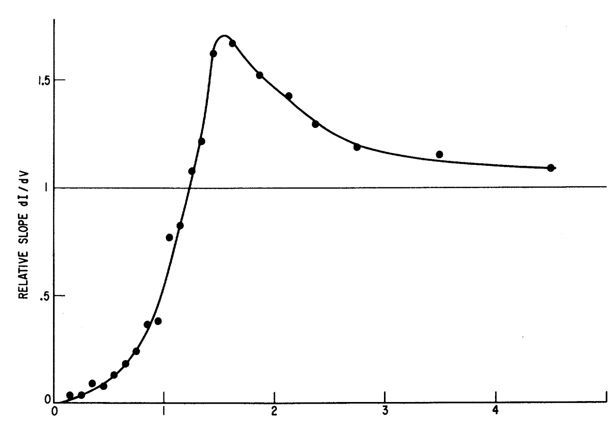

FIG. 2. From Fig. 1, slope dI/dV of curve 5 relative to slope of curve 1.

is given by the point at which the relative slope $dI/dV=1$. On this basis the gap width for lead is $(4.2\pm0.1)kT_c$.

The experiment has been repeated with tin and indium giving entirely similar results; the gap in each case is approximately $4kT_c$. These results are of a preliminary nature, and experiments at lower temperatures will make them more precise.

I wish to thank C. P. Bean and J. C. Fisher for their interest and encouragement, and P. E. Lawrence for his help in performing the experiments.

[1]J. C. Fisher and I. Giaever (to be published).

[2]W. A. Harrison (private communication) has pointed out that the tunnel current is not proportional to the density of states except in the limiting case of a low density of states.

[3]J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. 108, 1175 (1957).

CRITICAL FIELD FOR SUPERCONDUCTIVITY IN NIOBIUM-TIN

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(Received July 20, 1960)

It is well known[1] that Nb_3Sn is a superconductor with a high critical temperature, 18°K. The measurements here reported show that it has also an exceptionally high critical field, about 70 000 oersteds at 4.2°K, necessary for the suppression of all superconductivity.

The material was prepared by melting together niobium and tin in the argon arc, and the button so obtained was formed by grinding into a rod about 2 cm long and 4 mm in diameter, with rounded ends. The magnetic moment per gram, σ_g, was measured by pulling the specimen from one search coil to another in a constant field, the two search coils being connected in series opposition to a ballistic galvanometer. Calibration was with nickel of high purity.

Measurements were made in increasing fields, after cooling in zero field to liquid helium temperature. Results are shown in Fig. 1. The initial points (circles) follow accurately the line for $B=0$ ($H=-4\pi\sigma_g d$, where d is the density, 8.9), and then begin to deviate at about 4000 to 5000 oersteds. The variations in the readings in fields from 5000 to 20 000 oersteds reflect the well-known irregular changes in magnetization resulting from changes in domain structure in the intermediate state, as observed by Schawlow et al.[2] and others. The general shape of the magnetization curve is that observed in a hard superconductor. Polishing, or annealing the specimen at 1100°C for several hours, made no essential change in the character of the curve.

When the field was decreased from its maximum value (points marked with squares) some of the flux was frozen in, and irregularities were again observed.

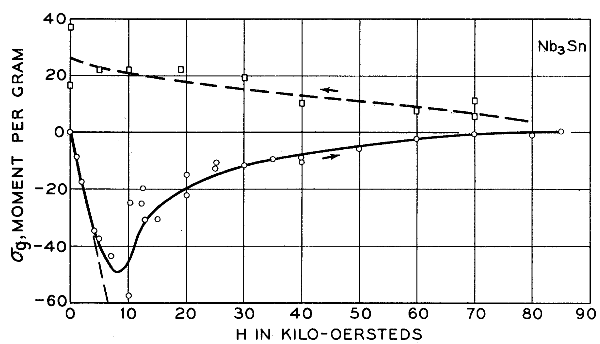

FIG. 1. Magnetization of Nb_3Sn as dependent on field strength, showing superconduction in entire specimen to about 5000 oersteds and superconduction in some parts of specimen to about 70 000 oersteds.

The authors are indebted to E. Corenzwit for preparation of the material, to W. E. Henry of the Naval Research Laboratory for details of the method of measurement, and to H. W. Dail for assistance with the experiment. The field was produced in a Bitter coil excited with a motor generator with a nominal power rating of one megawatt.

[1]B. T. Matthias and T. H. Geballe, Phys. Rev. 95, 1435 (1954).

[2]A. L. Schawlow, G. E. Devlin, and J. K. Hulm, Phys. Rev. 116, 626 (1959).

Study of Superconductors by Electron Tunneling

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If a small potential difference is applied between two metals separated by a thin insulating film, a current will flow due to the quantum mechanical tunnel effect. For both metals in the normal state the current-voltage characteristic is linear, for one of the metals in the superconducting state the current voltage characteristic becomes nonlinear, and for both metals in the superconductive state even a negative-resistance region is obtained. From these changes in the current voltage characteristics, the change in the electron density of states when a metal goes from its normal to its superconductive state can be inferred. By using this technique we have found the energy gap in metal films 1000–3000 A thick at 1°K to be $2\epsilon_{\mathrm{Pb}}=(2.68\pm0.06)\times10^{-3}$ ev, $2\epsilon_{\mathrm{Sn}}=(1.11\pm0.03)\times10^{-3}$ ev, $2\epsilon_{\mathrm{In}}=(1.05\pm0.03)\times10^{-3}$ ev, and $2\epsilon_{\mathrm{Al}}=(0.32\pm0.03)\times10^{-3}$ ev.

The variation of the gap width with temperature is found to agree closely with the Bardeen-Cooper-Schrieffer theory. Furthermore, the energy gap in these films has been found to depend upon the applied magnetic field, decreasing with increasing field.

INTRODUCTION

THE existence of an energy gap in superconductors is well documented experimentally, and is firmly grounded in the theory of superconductivity of Bardeen, Cooper, and Schrieffer.[1] Experimental evidence for the existence of a gap and, indirectly, its width, can be obtained from measurements of specific heat, thermal conductivity, nuclear relaxation, ultrasonic attenuation, and electromagnetic absorption.[2] In general, the width of the gap is inferred from the variation of one of the above parameters and, with the exception of electromagnetic absorption, represents only an indirect measurement.

This paper describes a method for investigating the energy gap and density of electron states in superconductors by means of electron tunneling through thin insulating films. It represents an entirely new approach to the problem and results in clear, unambiguous measurements of the energy gap. Some preliminary results, employing this method, have already been published.[3–6]

The samples used in this experiment consist of a thin insulating oxide layer sandwiched between two evaporated metal films. Experimentally, the electron tunneling current through the insulating oxide layer is observed as a function of the voltage applied between the two metal films. Because of their small physical size, the samples are well suited for standard low-temperature techniques.

If a *small* potential difference is applied to the two metals in their normal, nonsuperconducting state, the tunneling current through the insulating film will vary linearly with applied voltage, as long as the density of electron states in the two metals is constant over the applied voltage range.[7] On the other hand, if the density of electron states varies rapidly in this voltage range, as it does in superconductors, the current-voltage characteristics will be nonlinear. It appears that this nonlinearity is simply correlated with the variation in the density of electron states. In particular, no electrons can flow into the energy region of the gap in superconductors.

By this method we have measured the energy gap in lead, tin, indium, and aluminum. The variation of the energy gap as a function of temperature[6] and magnetic field has also been investigated.

APPARATUS

The apparatus which is shown in Fig. 1 consists basically of a liquid helium Dewar with provisions for

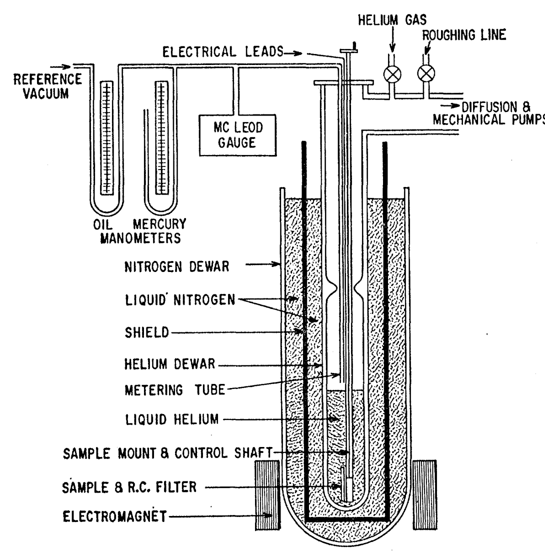

FIG. 1. A schematic drawing of the apparatus. The shield can be removed when studies are made using a magnetic field.

[1] J. Bardeen, L. N. Cooper, and J. R. Schrieffer, Phys. Rev. **108**, 1175 (1957).
[2] M. A. Biondi, A. T. Forrester, M. P. Garfunkel, and C. B. Satterthwaite, Revs. Modern Phys. **30**, 1109 (1958).
[3] I. Giaever, Phys. Rev. Letters **5**, 147 (1960).
[4] I. Giaever, Phys. Rev. Letters **5**, 464 (1960).
[5] J. Nicol, S. Shapiro, and P. H. Smith, Phys. Rev. Letters **5**, 461 (1960).
[6] I. Giaever, Proceedings of the Seventh International Conference on Low-Temperature Physics, Toronto, 1960 (to be published).
[7] J. C. Fisher and I. Giaever, J. Appl. Phys. **32**, 172 (1961).

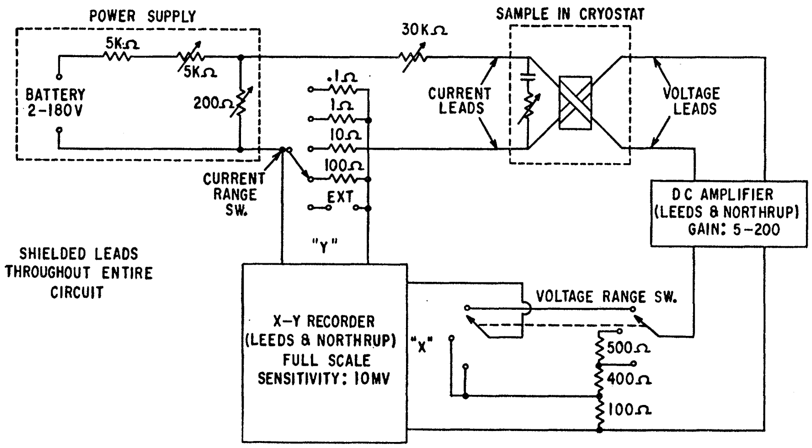

FIG. 2. Circuit diagram of the measuring circuit.

pumping on the helium, and an outer Dewar containing liquid nitrogen which acts as a radiation shield for the helium. The helium Dewar has a constriction in its diameter to minimize creep losses of the superfluid helium when the temperature is below the λ point.

Temperatures are measured by means of the helium vapor pressure. A metering tube which extends to within a few inches of the liquid helium level is connected to both oil and mercury manometers and to a McLeod gauge. The system is capable of attaining a temperature of about 0.9°K. Due to the low heat leakage, this temperature can be maintained for approximately six hours, which is adequate for making numerous measurements.

The electrical circuitry is shown in Fig. 2. To trace out the current-voltage characteristics, a Leeds and Northrup X-Y recorder and matching Leeds and Northrup dc amplifier are used in conjunction with external shunts and multipliers to extend the range of the instruments. These instruments contain chopper-stabilized amplifiers and, therefore, have virtually no drift. To accommodate various sample resistances and to obtain the necessary detail in the current-voltage curves, the current scale can be decade switched over a full-scale sensitivity from 100 ma to 1 μa and the voltage scale from 100 mv to 50 μv.

The emf source can be used as either a high- or low-impedance source, by suitable adjustment of the two variable resistors and the applied battery voltage. The high capacitance of the sample in conjunction with considerable lead inductance gives rise to very troublesome high-frequency oscillations whenever the sample is biased into its negative-resistance region. By placing an adjustable high-pass filter in parallel with the sample, the high-frequency oscillations can be eliminated or at least greatly reduced. The high-pass filter consists of a capacitor large in comparison to the sample capacitance and a variable resistor in series, and is in close proximity to the sample to minimize lead inductance. The oscillations are reduced by matching the variable resistance to the negative resistance of the sample. The variable resistor is a Bourns Trimpot which is mounted on the end of a $\frac{1}{4}\times 0.008$ in. stainless steel tube. Concentric with this tube is a $\frac{1}{8}\times 0.008$ in. stainless steel tube which engages the adjustment screw of the resistor and passes through an O-ring seal in the Dewar cover plate, to permit external adjustments.

The electrical connections into the Dewar, consisting of current and voltage leads, are brought out through the cover plate and are sealed in place with Apiezon wax to achieve a tight seal. In order to minimize heat leaks, four 3-mil Formex-covered copper wires are used inside the cryostat. To minimize induced noise, the entire electrical circuitry outside the Dewar is shielded. The sample and leads within the Dewar can be shielded by a copper-clad soft iron shield which sits in the liquid nitrogen, surrounding the helium Dewar. This shield is not used for measurements made with an externally applied magnetic field.

Since the voltages applied to the sample are very small, induced voltages caused by ever-present fluctuating stray fields remain a difficult problem even after careful shielding. The slow response time of the recording apparatus causes the readings to be averaged over the fluctuations resulting from induced voltages. Due to the nonlinear characteristics of our samples, this averaging tends to smooth out the current-voltage curves and results in lost detail. The difficulty is virtually eliminated by including a resistance in series with the current loop, and making this resistance as large as practical. The large resistance in series with the sample resistance acts as a voltage divider for induced noise, so that only a small amount of noise appears across the sample. This large series resistance effectively increases the emf source impedance and cannot be used when investigating the negative-resistance region of the sample. It has, however, been retained for measurements outside the negative resistance region on most samples.

The sample is mounted directly on the variable resistor which is a part of the high-pass filter. This insures mechanical rigidity and reproducible, accurate positioning for measurements involving magnetic fields.

When the sample is subjected to a magnetic field, it is aligned so that the field vector is in the plane of both films, parallel to the long dimension of the aluminum film, and normal to the long dimension of the other metal film.

SAMPLE PREPARATION

The sample consists of two metal films separated by a thin insulating layer. Aluminum/aluminum oxide/metal sandwiches are prepared by vapor-depositing aluminum on microscope glass slides in vacuum, oxidizing the aluminum, and then vapor-depositing a metal over the aluminum oxide. First the microscope slide is cut to size, $\frac{1}{2}\times 3$ in., so as to fit through the constriction in the helium Dewar. Next, indium is smeared onto the four corners of the glass slide to provide contacts between the evaporated metal strips and external leads. The glass slide with indium contacts is then washed with Alconox detergent, rinsed with distilled water and ethanol, and dried with dry nitrogen gas.

Next, the glass slide is mounted in the evaporator so that it can be positioned behind suitable masks in vacuum. The evaporations are made from tantalum strips approximately $\frac{3}{8}\times 1\frac{1}{2}\times 0.005$ in., which have previously been charged and heated in vacuum so that the charge wets the tantalum strip. The evaporations are made at a starting pressure of 5×10^{-5} mm Hg, or less.

Preparation of the metal/insulator/metal sandwich proceeds in three distinct steps, as shown in Fig. 3, during which the substrate is at room temperature.

First, a layer of aluminum is evaporated onto the glass slide between two contacts. This strip is 1 mm wide and 1000–3000 A thick. Next, the aluminum is oxidized either at atmospheric pressure or some reduced pressure. Finally, a layer of Al, Pb, In, or Sn, of dimensions similar to the aluminum strip, is evaporated over the aluminum oxide layer between the remaining two contacts.

The thickness of the Al_2O_3 insulating layer between the metal strips is subject to a number of variables. The pressure and time dependence of oxidation rate has been extensively investigated and is well documented in the literature.[8] Atmospheric humidity or residual H_2O vapor in the vacuum system also affects the oxidation rate. We have found that an increase in the amount of water effects a more rapid oxide growth rate. The oxide thickness is also contingent upon the evaporation rate and evaporation temperature of the metal layer deposited over the oxide layer. Presumably, metals which require higher temperatures for evaporation must give up more energy to the oxide film; i.e., the atoms penetrate further into the oxide, thereby effectively reducing the thickness of the oxide layer. Another parameter is oxidation temperature, elevated temperatures promoting an increase in the oxidation rate. A final variable is introduced by the evaporation rate of the aluminum layer, which influences its surface characteristics. We have noted that the oxide grows more slowly and reaches a thinner limiting value on films which were evaporated with a high deposition rate (approximately 1000 A/sec).

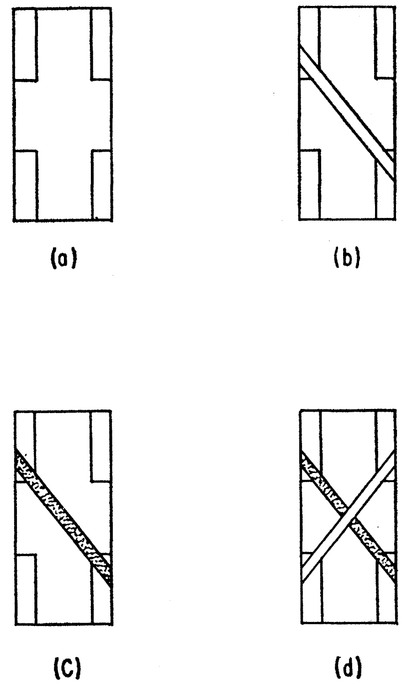

FIG. 3. Sample preparation. (a) Glass slide with indium contacts. (b) An aluminum strip has been deposited across the contacts. (c) The aluminum strip has been oxidized. (d) A lead film has been deposited across the aluminum film, forming an $Al\text{-}Al_2O_3\text{-}Pb$ sandwich.

TABLE I. Approximate relationship between oxidation time and film resistance for $Al\text{-}Al_2O_3\text{-}Pb$ sandwiches.

Time	Temperature	Pressure	Resistance (ohm/mm^2)
24 hr	100°C	atmospheric	10^5-10^7
24 hr	room	atmospheric	10^3-10^5
10 min	room	atmospheric	$10-10^3$
2 min	room	atmospheric	$1-10^2$
10 min	room	200μ Hg	10^1-10^{-1}
10 min	room	50μ Hg	$10^{-2}-10^{-1}$

By controlling these parameters to some extent, the resistance of a 1-mm^2 junction can be made to vary between 10^{-2} and 10^7 ohm. Typical oxidation conditions for an $Al\text{-}Al_2O_3\text{-}Pb$ sandwich are given in Table I. (The resistance variations are probably due to the effect of humidity and the surface characteristics of the aluminum layer.)

It is possible to measure indirectly the thickness of the oxide layer by measuring the capacitance of the junction and then calculating the thickness.[7]

MODEL

The concept that particles can penetrate energy barriers is as old as quantum mechanics. In nuclear

[8] D. D. Eley and P. R. Wilkinson, *Structure and Properties of Thin Films* (John Wiley & Sons, Inc., New York, 1959), p. 508.

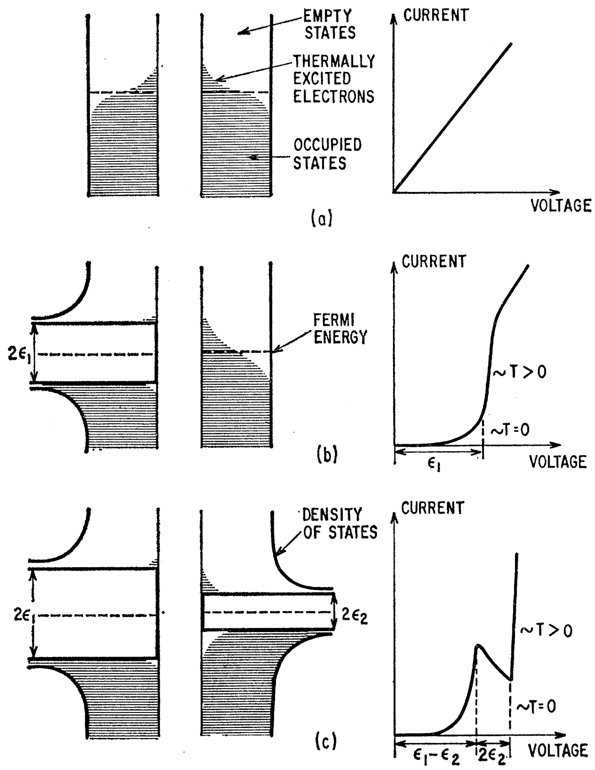

FIG. 4. Energy diagram displaying the density of states and the current-voltage characteristics for the three cases. (a) Both metals in the normal state. (b) One metal in the normal state and one in the superconducting state. (c) Both metals in the superconducting state.

physics, for example, the theory of α decay depends upon this tunnel effect. It has long been known that an electric current can flow between two metals separated by a thin insulating film because of the quantum-mechanical tunnel effect. Theoretical calculations were first made by Sommerfeld and Bethe[9] for a small potential difference applied between the two metals. These calculations were later extended by Holm.[10] An important result of these calculations is that for small voltages across the insulating film the tunnel current through the film is proportional to voltage. Holm *et al.*[11] furnished early experimental evidence for the tunneling effect, a work which was extended by Dietrich[12] and later by Fisher and Giaever.[7] The experiment of tunneling into superconductors[3] furnishes unquestionable evidence that this conduction mechanism is responsible for practically the whole current flow. In the following discussion we shall treat only the low-voltage region where the current flowing through the insulating film is proportional to the voltage across the film, provided both superconductors are in the normal state.

In Fig. 4(a) we show a simple model of two metals separated by a thin insulating film, the insulating film is pictured as a potential barrier. In Fig. 4(b) is shown the case when one of the two metals is in the superconducting state. Note how the electron density of states has changed, leaving an energy gap centered at the Fermi level as postulated by Bardeen, Cooper, and Schrieffer.[1] This particular model of a superconductor is a one-particle approximation, and it gives a surprisingly accurate picture of the experiments. In Fig. 4(c) both the metals are pictured in the superconducting state.

First we shall discuss qualitatively these three different cases, and later quantitatively calculate the current when both metals are in the normal state, and when only one of the metals is in the normal state.

The transmission coefficient of a quantum particle through a potential barrier depends exponentially upon the thickness of the barrier and upon the square root of the height of the barrier. For small voltages applied between the two metals neither the barrier thickness nor the barrier height is altered significantly. The current will then be proportional to the applied voltage, because the number of electrons which can flow increases proportionally to the voltage. The temperature effect will be very small, as the electron distribution is equal on either side of the barrier with metals in the normal state and in addition, kT is much smaller than the barrier height.

When one of the metals is in the superconducting state the situation is radically different. At absolute zero temperature, no current can flow until the applied voltage corresponds to half the energy gap. Assuming that the current is proportional to the density of states, the current will increase rapidly with voltage at first, and then will asymptotically approach the current-voltage characteristic found when both metals were in the normal state. At a temperature different from zero we will have a small current flow even at the lowest voltages. But since the two sides of the barrier now look different, the current will depend strongly upon temperature.

When both metals are in the superconducting state, the situation is again different. At absolute zero no current can flow until the applied voltage corresponds to half the sum of the two energy gaps. At a finite temperature, a current again will flow at the smallest applied voltages. The current will increase with voltage until a voltage equal to approximately half the difference of the two energy gaps is applied. When the voltage is increased further it is possible for only the same number of electrons to tunnel, but since the electrons will face a less favorable (lower) density of states, the current will actually decrease with increasing voltage. Finally, when a voltage equal to half the sum of the two gaps is applied the current will again increase rapidly with voltage and approach asymptotically the

[9] A. Sommerfeld and H. Bethe, *Handbuch der Physik*, edited by S. Flügge (Verlag Julius Springer, Berlin, 1933), Vol. 24, Part 2, p. 333.
[10] R. Holm, J. Appl. Phys. **22**, 569 (1951).
[11] R. Holm, *Electric Contacts* (Hugo Geber, Stockholm, Sweden, 1946).
[12] I. Dietrich, Z. Physik **132**, 231 (1952).

current-voltage characteristics obtained when both metals were normal.

Since we regard the distributions of holes and electrons in the metals in both the normal and superconducting state as symmetric about the Fermi level, no rectification effects are expected.

If we regard the tunneling through the insulating layer as an ordinary quantum-mechanical transition, the transition probability from an occupied state $\mathbf{k}$ on the left side of the barrier to a state $\mathbf{k}'$ on the right side can be written:

$$P_{\mathbf{k}\to\mathbf{k}'} = (2\pi/\hbar)|M|^2 n'(1-f'), \tag{4.1}$$

where n' is the density of states on the right side and f' the probability that the state $\mathbf{k}'$ is occupied. $|M|^2$ is the matrix element for the transition and we assume $|M|^2$ vanishes unless the components of $\mathbf{k}$ and $\mathbf{k}'$ transverse to the boundary are equal; that is, specular transmission and then n' is the density of states on the right for fixed wave number components parallel to the boundary. This is a convenient though not an essential assumption.

To calculate the current from left to right we sum over occupied states on the left and obtain

$$i = (4\pi e/\hbar)\textstyle\sum_{k_t}\sum_{k_x}|M|^2 n' f(1-f'), \tag{4.2}$$

where k_t is the component of wave number transverse to the barrier, k_x the component perpendicular to the barrier, e the electron charge, and f the probability that state $\mathbf{k}$ is occupied.

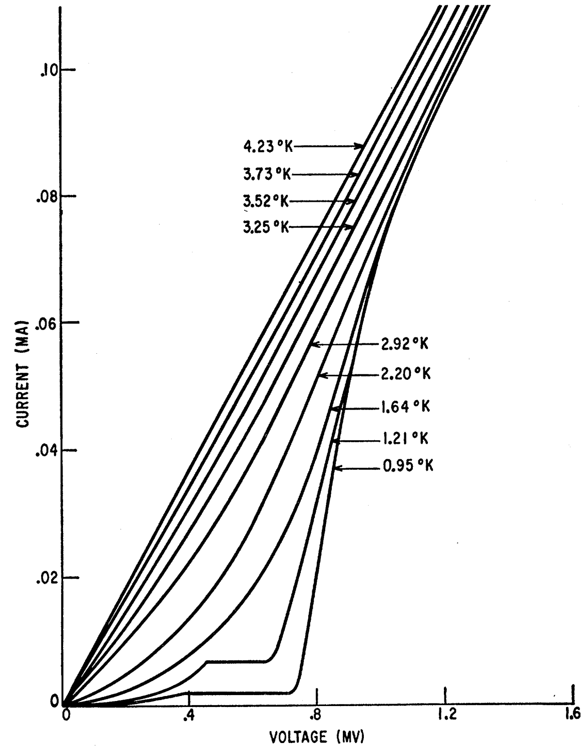

FIG. 6. Current-voltage characteristics of an Al-Al_2O_3-Sn sandwich at various temperatures.

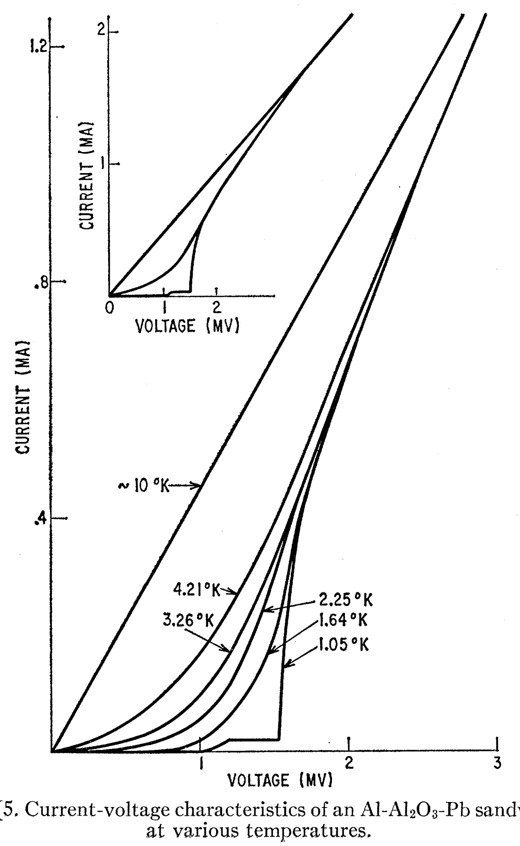

FIG. 5. Current-voltage characteristics of an Al-Al_2O_3-Pb sandwich at various temperatures.

By converting the sum over k_x to an integral over energy with fixed k_t we get

$$i = A\sum_{k_t}\int_{-\infty}^{\infty}|M|^2 nn' f(1-f')dE, \tag{4.3}$$

where A is a constant, n the density of states at the left side of the barrier (for fixed k_t), and E the energy measured from the Fermi energy.

By subtracting a similar expression for the current flowing from right to left, we get the net current flow:

$$I = A\sum_{k_t}\int_{-\infty}^{\infty}|M|^2 n' n(f-f')dE. \tag{4.4}$$

To fit the experimental results it is necessary to assume that $|M|^2 \approx$ constant. Bardeen,[13] using a many-particle point of view in connection with the WKB method, finds it plausible that $|M|^2$ is a constant over the energy values of interest. On assuming a constant $|M|^2$ and spherical symmetry of the dependence of energy on wave number, n' and n which are one dimensional densities of states are proportional to the total densities of states, and we may therefore sum over k_t directly and take $|M|^2$ out of the integral. We are

[13] J. Bardeen, Phys. Rev. Letters **6**, 57 (1961).

left with

$$I = A' \int_{-\infty}^{\infty} n'n\{f(E) - f(E+eV)\} dE, \tag{4.5}$$

where A' is a constant and eV is the difference between the two Fermi levels. (V is the applied voltage.)

For the current between two normal metals we obtain at absolute zero and for small applied voltages:

$$I_{NN} = A'n'(E_F)n(E_F)eV, \tag{4.6}$$

i.e., the current is proportional to voltage.

For a superconductor we may take the density of states from the Bardeen-Cooper-Schrieffer theory:

$$n_s = n\frac{E}{(E^2-\epsilon^2)^{\frac{1}{2}}}, \tag{4.7}$$

where E is measured from the Fermi energy, and ϵ is half the energy gap. Thus the current between one metal in the normal state and one metal in the superconducting state can be written:

$$I_{NS} = A'n'(E_F)n(E_F)\int_{-\infty}^{\infty} \frac{|E|}{(E^2-\epsilon^2)^{\frac{1}{2}}} \times [f(E) - f(E+eV)] dE. \tag{4.8}$$

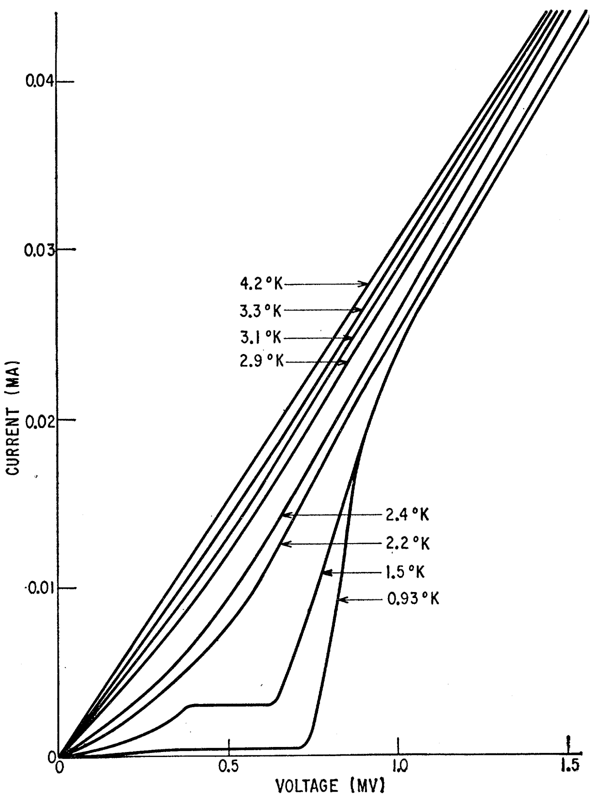

FIG. 7. Current-voltage characteristics of an Al-Al_2O_3-In sandwich at various temperatures.

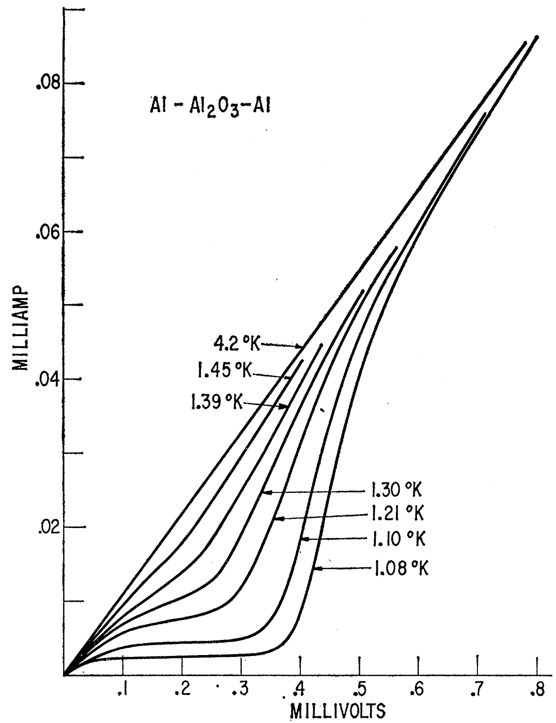

FIG. 8. Current-voltage characteristics of an Al-Al_2O_3-Al sandwich at various temperatures.

For small applied voltages such that $eV < \epsilon$ we may evaluate the above integral, as shown in Appendix I, and obtain:

$$I_{NS} = 2C_{NN}\frac{\epsilon}{e}\sum_{m=1}^{\infty}(-1)^{m+1}K_1\left(m\frac{\epsilon}{kT}\right)\sinh\left(m\frac{eV}{kT}\right), \tag{4.9}$$

where C_{NN} is the conductance when both metals are in the normal state, K_1 is the first order of the modified Bessel function of the second kind, e the electron charge, k the Boltzmann constant, T the temperature, and m an integer. Evaluation of (4.9) for special cases is given in Sec. (c) below. Calculations of the current for $eV > \epsilon$ and for tunneling between two superconductors, require more extensive computation.

Finally, it should be mentioned here that we have treated the insulating layer as if it were a vacuum. However, since the insulator has both a conduction band and a valence band, we could possibly also get a "hole" current. In this particular case this is of little importance as we are mostly interested in the current ratio I_{NS}/I_{NN} rather than the absolute values of current.

EXPERIMENTAL RESULTS

(a) Energy Gaps

We report on four different combinations of superconductors namely Al-Al_2O_3-Pb, Al-Al_2O_3-Sn, Al-Al_2O_3-In,

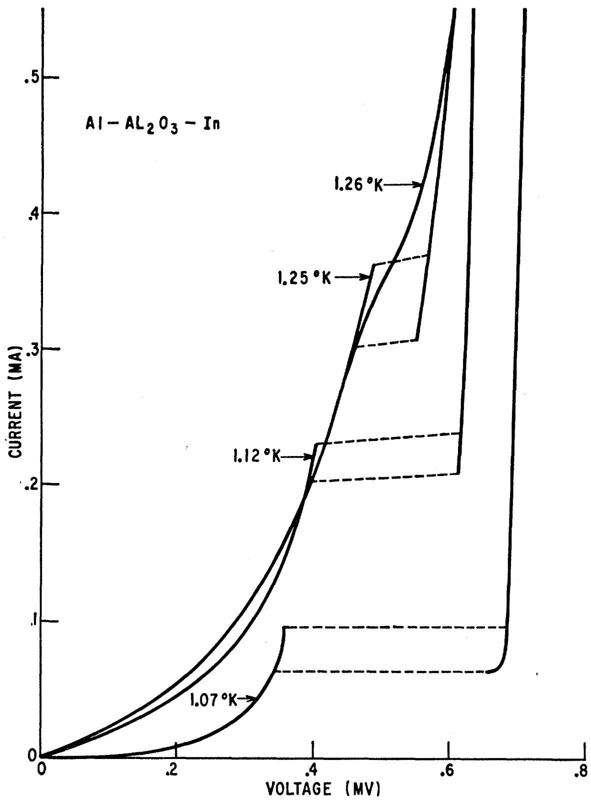

FIG. 9. Detailed current-voltage characteristics of an Al-Al_2O_3-In sandwich, showing the change of energy gap in Al as a function of temperature.

and Al-Al_2O_3-Al. The current-voltage characteristics at various temperatures for these four systems are shown in Figs. 5, 6, 7 and 8, respectively. As seen, the general behavior of the current-voltage characteristics is as predicted from the model. The negative resistance regions are not very apparent on these curves due to the current scale chosen. When the energy gaps on either side of the barrier are equal, as is the case for the Al-Al_2O_3-Al sandwich, a negative resistance region should be observable as well at sufficiently low temperatures. We did not observe this, however, due to temperature limitations in the experimental setup. Note in particular the insert on Fig. 5, showing that for larger voltages the current-voltage characteristics are independent of whether the metals are in the normal or superconducting states. This fact strongly supports the assumption that the tunnel current between superconductors is proportional to the density of states.

From these curves we find the energy gaps at approximately 1°K:

$$2\epsilon_{Pb}=(2.68\pm0.06)\times10^{-3}\text{ ev}=(4.33\pm0.10)kT_c,$$
$$2\epsilon_{Sn}=(1.11\pm0.03)\times10^{-3}\text{ ev}=(3.46\pm0.10)kT_c,$$
$$2\epsilon_{In}=(1.05\pm0.03)\times10^{-3}\text{ ev}=(3.63\pm0.10)kT_c,$$
$$2\epsilon_{Al}=(0.32\pm0.03)\times10^{-3}\text{ ev}=(3.20\pm0.30)kT_c.$$

While the energy gaps in Pb, Sn, and In will not change significantly between 1° and 0°K, this is not true for Al, because of its low transition temperature. It should be noted that the transition temperature for the aluminum films varied from sample to sample, was always greater than the bulk transition temperature, and increased with decreasing thickness of the aluminum films. The highest transition temperature observed for Al was 1.8°K. In calculating the energy gaps in terms of kT_c the bulk transition temperature has been used for all films. One reason for this choice is that the observed energy gap in the aluminum films at 1°K is approximately 0.32×10^{-3} ev, regardless of the transition temperatures observed. This experimental result may be due to the broad transition region usually observed in evaporated films.

(b) Variation of the Energy Gap with Temperature

In Fig. 9 we show detailed current-voltage characteristics for an Al-Al_2O_3-In sandwich as a function of temperature. Because the curves are traced out using a constant current source rather than a constant voltage source, the negative resistance region appears as a hysteresis loop. The width of this loop corresponds ap-

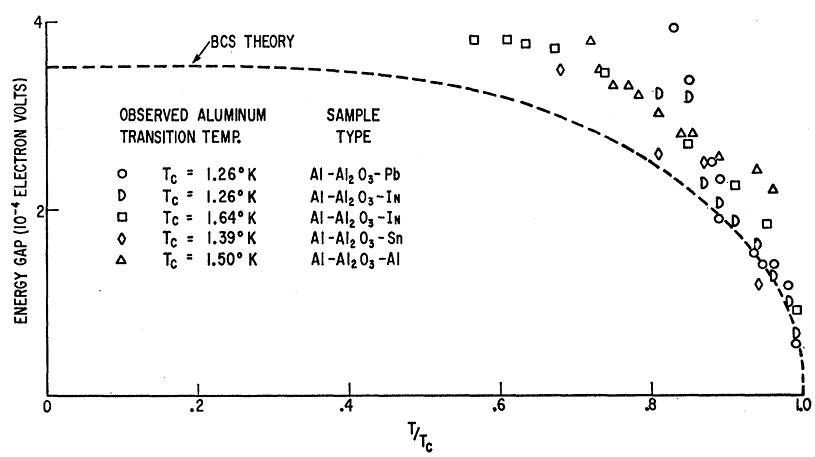

FIG. 10. The energy gap as a function of reduced temperature for several aluminum films, compared with the Bardeen-Cooper-Schrieffer theory.

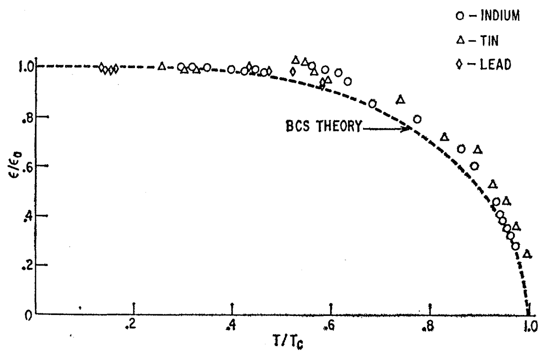

FIG. 11. The energy gap of Pb, Sn, and In films as a function of reduced temperature, compared with the Bardeen-Cooper-Schrieffer theory.

proximately to the full gap width in aluminum, and we can clearly see the variation of gap width with temperature. In Fig. 10 we have plotted the variation of the gap width as a function of reduced temperature for several different samples. For this figure we have used the observed value of the transition temperature T_c. As seen from the figure, the energy gap at $T=0$ does not appear to be very sensitive to the variations in the transition temperature actually observed for the aluminum films. One reason for this could be that the whole area of the aluminum film does not become superconducting at the same temperature, due to localized stresses or impurities. The best estimate of the energy gap for aluminum at absolute zero is

$$2\epsilon_{\mathrm{Al}}=(4.2\pm0.6)kT_c=(0.42\pm0.06)\times10^{-3}\ \mathrm{ev},$$

where T_c is taken as the bulk value.

It is possible to observe directly the variation of the energy gap in aluminum over the entire applicable temperature range. The energy gap in indium, tin, and lead can also be observed directly in the temperature range in which aluminum is superconducting. At higher temperatures the gap in lead, tin, and indium is not directly observable; however, we are able to calculate the gap width for all temperatures. By letting $V\rightarrow0$ in Eq. (4.9), we may write:

$$\frac{I_{NS}}{I_{NN}}=2\sum_{m=1}^{\infty}(-1)^{m+1}m\frac{\epsilon}{kT}K_1\left(m\frac{\epsilon}{kT}\right). \quad (5.1)$$

The quality I_{NS}/I_{NN} as $V\rightarrow0$ is easily obtained from the experimental results and we may then calculate ϵ from Eq. (5.1). The results are shown in Fig. 11 and are in good agreement with the theory. It should be pointed out that the values of the energy gap, calculated in this way, are in agreement with the directly observed values in the temperature range where both of these measurements can be made. This is most gratifying since these measurements are independent of each other, one being defined at absolute zero, and the other arising solely from the temperature-dependence of the current. The calculated values of the energy gap may appear somewhat too large at low temperatures for some samples due to noise in the measuring circuit.

(c) Calculated versus Measured Current

For tunneling between a metal in the normal state and a metal in the superconducting state, we again use Eq. (4.9) and restrict the calculations to the region where $\epsilon>eV$. In Fig. 12, we compare the calculated values of current with the experimental results obtained on an Al-Al_2O_3-Pb sandwich at various temperatures. The agreement is very good using only two terms of the series in Eq. (4.9). Note in particular that for $\epsilon\gg kT$ and for large voltages such that $\sinh(eV/kT)\approx\frac{1}{2}\exp(eV/kT)$, we may write

$$\ln I_{NS}=\frac{1}{kT}eV+\alpha(\epsilon,T), \quad (5.2)$$

where α is some function of ϵ and T, independent of V. Thus we can determine the temperature directly from the slope when we plot $\ln I_{NS}$ versus V.

(d) Variation of the Energy Gap with Magnetic Field

By subjecting these samples to a magnetic field parallel to the plane of the metal films, we have found that the energy gap is a function of the applied field. In Fig. 13 we show some detailed results obtained on an Al-Al_2O_3-Pb sandwich. These results are summarized in

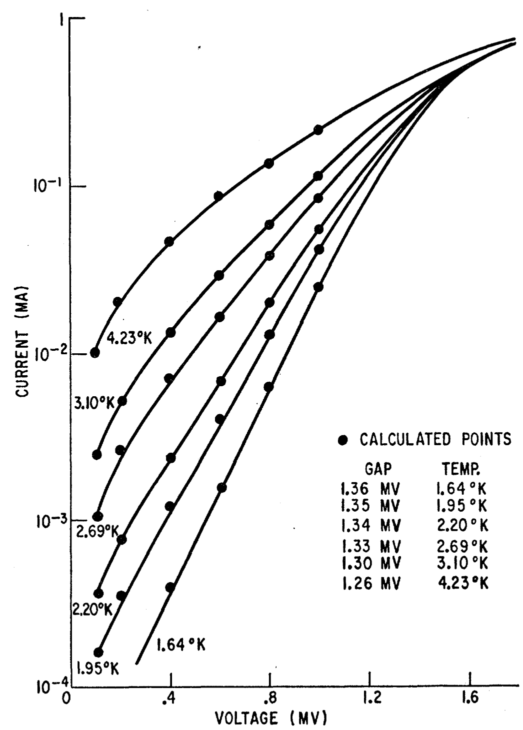

FIG. 12. Observed current-voltage characteristics for an Al-Al_2O_3-Pb sandwich at various temperatures, versus calculated values using the Bardeen-Cooper-Schrieffer density of states.

Fig. 14 where the gap width for aluminum is shown as a function of magnetic field. This curve does not agree with the observed fact that for bulk materials the transition between the normal and superconducting state is a first-order transition. A first-order transition would require a discontinuous change in gap width at the critical field. While this discrepancy may arise from the possibility that the transition is not of first order in a thin film, we believe it more likely that the surface roughness of the film will cause the magnetic field to be nonuniform. This nonuniformity will tend to smear the discontinuous change in gap that we expect at the critical field.

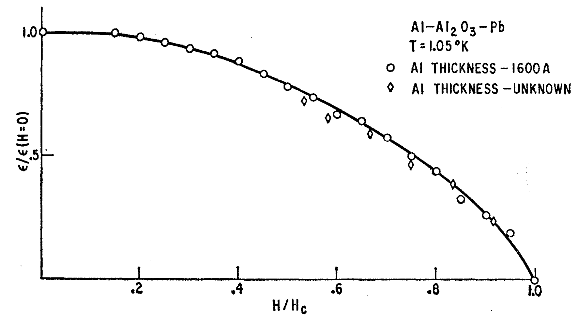

FIG. 14. Apparent variation of the energy gap in an aluminum film as a function of the applied magnetic field.

To make sure that the change in the current-voltage characteristics is due to a change in the energy gap, rather than being due to the aluminum film going into the intermediate state, we also investigated the effect of the magnetic field on the energy gap of lead. In Fig. 15 we plot current versus voltage for an Al-Al_2O_3-Pb sandwich at various magnetic fields. If we deal with the intermediate state in lead, then the observed current should be the sum of a current varying linearly with voltage and a current varying exponentially with voltage. This is clearly not so. On the other hand, a good fit to these curves can be obtained by using the expression derived for the tunnel current between one normal and one superconducting member with a varying gap for different field strengths. To obtain a good fit, it is necessary to use a rather large gap. This is probably due to noise in the measuring circuit or possibly a nonuniform energy gap in lead. It should be mentioned, although no detailed investigation has been made by us, that for thinner films much higher fields are needed to observe the change in the energy gap.

(e) Density of States

The good agreement between the experimental and calculated currents, using the density of states from the

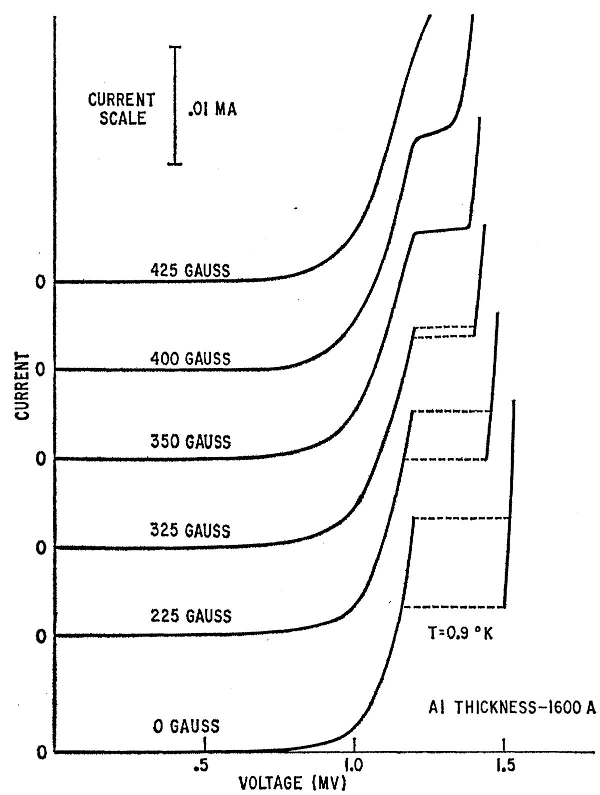

FIG. 13. Detailed current-voltage characteristics of an Al-Al_2O_3-Pb sandwich, showing a change in the energy gap of aluminum as a function of the applied magnetic field.

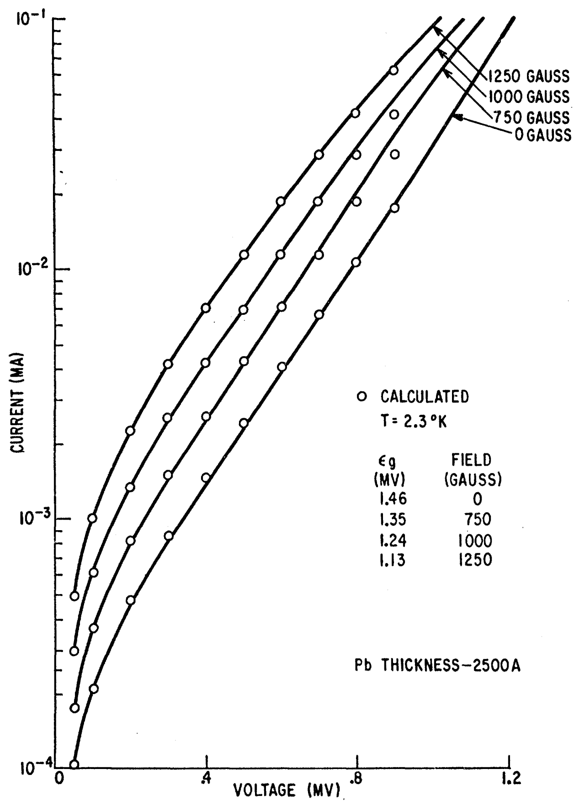

FIG. 15. The change in the current-voltage characteristics of an Al-Al_2O_3-Pb sandwich as a function of the magnetic field, demonstrating that the observed change cannot be due to the lead film being in the intermediate state.

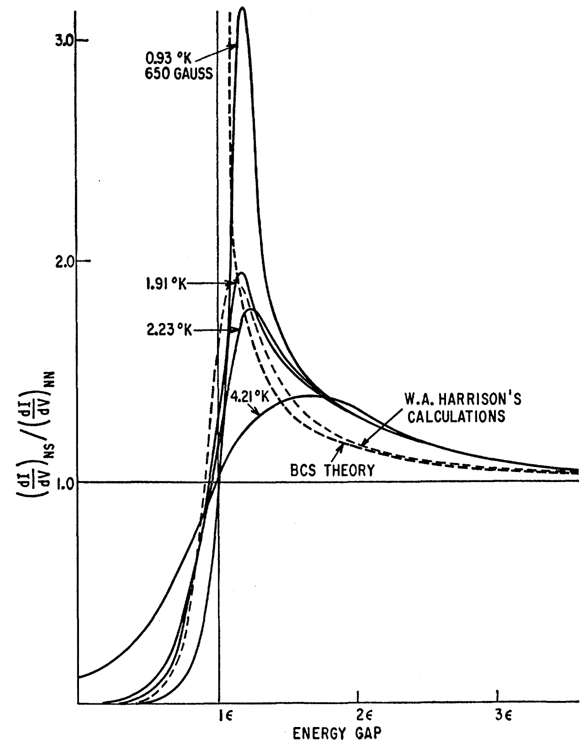

FIG. 16. The relative conductance for an Al-Al_2O_3-Pb sandwich, i.e., the conductance of the sandwich when the lead film is in the superconducting state, divided by the conductance when the lead film is in the normal state, plotted against energy, and compared with the Bardeen-Cooper-Schrieffer density of states. This density of states is used by W. Harrison in his calculations, with $\epsilon/kT=10$.

Bardeen-Cooper-Schrieffer theory, is a great triumph for this theory. In deriving Eq. (4.9), we have integrated over the density of states so that the current is relatively insensitive to small variations in the density of states. Under the assumption that the current is proportional to the density of states, we should get the relative change in the density of states directly by plotting the conductance when one of the metals is superconductive $(dI/dV)_{NS}$ divided by the conductance when both metals are normal $(dI/dV)_{NN}$ against energy. In Fig. 16 we show these results, obtained from an Al-Al_2O_3-Pb sandwich at four different temperatures. Note that at the lowest temperature we have kept the aluminum normal by applying a magnetic field and this again smears the energy gap in lead, making it difficult to assign a specific value to the gap. We see that in spite of the kT smearing, the density of states strongly resembles the theoretical density of states.

(f) Negative-Resistance Region

In spite of the damping RC network used in parallel with the sample, we found it difficult to eliminate self-induced oscillations in the negative-resistance region. In Fig. 17 we show two attempts to trace out the negative resistance region. We believe that induced noise in the measuring circuit is the limiting factor in tracing out the negative-resistance region, as literally microvolts of induced noise will smear out the curves.

(g) Effect of Metal Bridges and Trapped Flux

In Fig. 18 we show the effect of a metal bridge short-circuiting the sample. The bridge is initially superconducting so that no voltage can be applied across the sample. Then, at a certain current density the bridge becomes normal, but now its resistance is too large to appreciably affect the tunnel current. When the voltage

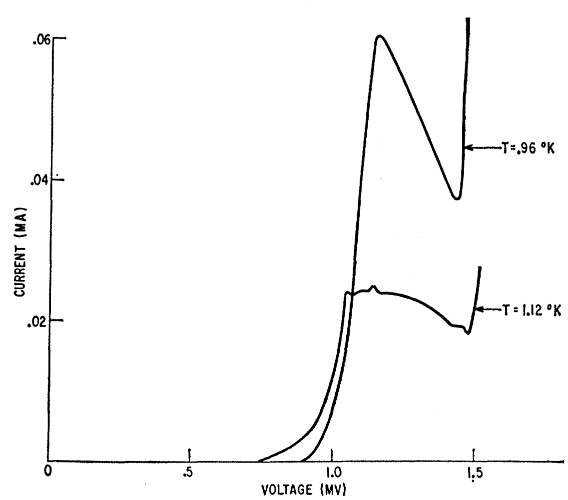

FIG. 17. The negative-resistance region traced out for two different Al-Al_2O_3-Pb sandwiches. We believe the wiggles in the lower curve are due to oscillations in the circuit.

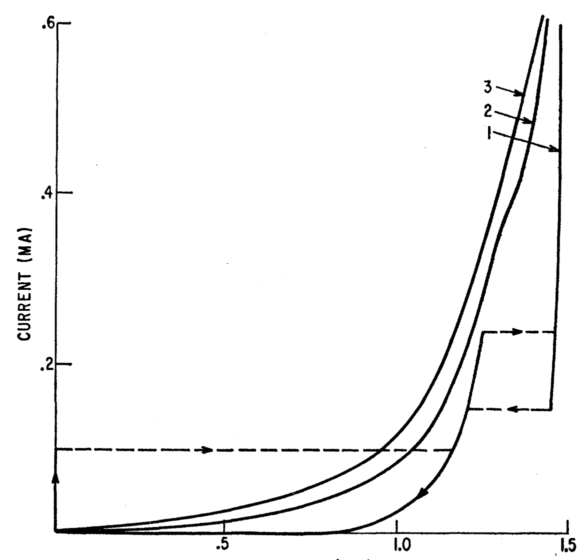

FIG. 18. The effect of trapped flux on the current-voltage characteristic of an Al-Al_2O_3-Pb sandwich. (1) The sample with no field applied; (2) the external field removed, showing the effect of the trapped flux. The figure also shows the effect of a metal bridge across the insulating film; (3) with a magnetic field applied normal to the surface of the films.

is again reduced, the bridge remains normal at a lower current density due to Joule heating.

In Fig. 18 we also show the effect of trapped flux, when the magnetic field purposely has been applied normal to the films. The trapped flux has a large effect upon the current-voltage characteristics, and this technique may possibly be helpful in studying the intermediate state.

(h) Other System

All the experiments we report on have been done by using Al_2O_3 as the insulating layer; however, the experiments may be done by using other insulating layers as well. For example, we have observed tunneling through tantalum and niobium oxides. In these experiments we used bulk specimens of tantalum and niobium; however, we did not observe any evidence for an energy gap in any of these materials. We believe the reason for this is that due to impurities, the surfaces of these materials did not become superconducting. Another superconductor used by us is lanthanum, in which we have observed evidence for an energy gap.

SUMMARY

The method of studying superconductors by electron tunneling has been very successful, and the results are in good agreement with the Bardeen-Cooper-Schrieffer theory. We have directly verified the change of energy gap with temperature. Also, we have shown that for thin films the energy gap is a function of the magnetic field.

ACKNOWLEDGMENTS

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APPENDIX I

To evaluate the expression:

$$I_{NS}=A'n'(E_F)n(E_F)\int_{-\infty}^{\infty}\frac{|E|}{(E^2-\epsilon^2)^{\frac{1}{2}}}\times[f(E)-f(E+eV)]dE, \quad \text{(A.1)}$$

we introduce the conductance C_{NN} when both metals are normal, i.e.,

$$\frac{I_{NN}}{V}=C_{NN}=A'n'(E_F)n(E_F)e, \quad \text{(A.2)}$$

and split the integral into two parts:

$$I_{NS}=\frac{C_{NN}}{e}\int_{+\epsilon}^{\infty}\frac{E}{(E^2-\epsilon^2)^{\frac{1}{2}}}\{f(E)-f(E+eV)\}dE - \frac{C_{NN}}{e}\int_{-\infty}^{-\epsilon}\frac{E}{(E^2-\epsilon^2)^{\frac{1}{2}}}\{f(E)-f(E+eV)\}dE. \quad \text{(A.3)}$$

By introducing $x+\epsilon=E$ in the first integral and $x+\epsilon=-E$ in the second integral, we get

$$I_{NS}=\frac{C_{NN}}{e}\int_{0}^{\infty}\frac{x+\epsilon}{[(x+2\epsilon)x]^{\frac{1}{2}}}[f(x+\epsilon)-f(x+\epsilon+eV)]dx + \frac{C_{NN}}{e}\int_{\infty}^{0}\frac{x+\epsilon}{[(x+2\epsilon)x]^{\frac{1}{2}}}\times\{f[-(x+\epsilon)]-f[eV-(x+\epsilon)]\}dx, \quad \text{(A.4)}$$

and because the Fermi function is an even function,

$$I_{NS}=\frac{C_{NN}}{e}\int_{0}^{\infty}\frac{x+\epsilon}{[(x+2\epsilon)x]^{\frac{1}{2}}}\times[f(x+\epsilon-eV)-f(x+\epsilon+eV)]dx. \quad \text{(A.5)}$$

By expanding the Fermi function in a series valid for $\epsilon>eV$, we obtain

$$I_{NS}=2\frac{C_{NN}}{e}\sum_m(-1)^{m+1}e^{-m(\epsilon/kT)}\sinh(meV/kT)\times\int_0^{\infty}\frac{x+\epsilon}{[(x+2\epsilon)x]^{\frac{1}{2}}}e^{-m(x/kT)}dx. \quad \text{(A.6)}$$

The last integral is of a known Laplace-integral form [A. Erdélyi, *Table of Integral Transforms* (McGraw-Hill Book Company, Inc., 1954)], and we obtain

$$I_{NS}=2C_{NN}\frac{\epsilon}{e}\sum_m(-1)^{m+1}K_1(m\epsilon/kT)\sinh(meV/kT), \quad \text{(A.7)}$$

where K_1 is the first-order modified Bessel function of the second kind.

APPENDIX II

Note added in proof. It is of interest to compare the values of the energy gaps obtained by using electron tunneling to previous direct measurements of the energy gap (Table II).

TABLE II.

Superconductor	Energy gap in units of kT_c: Tunneling measurements	Richards and Tinkham[a]	Ginsburg and Tinkham[b]
Indium	3.63±0.1	4.1±0.2	3.9±0.3
Tin	3.46±0.1	3.6±0.2	3.3±0.2
Lead	4.33±0.1	4.1±0.2	4.0±0.5

[a] P. L. Richard and M. Tinkham, Phys. Rev. 119, 581 (1960).
[b] D. M. Ginsburg and M. Tinkham, Phys. Rev. 118, 990 (1960).