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RF SUPERCONDUCTING MATERIALS RESEARCH AT SLAC*

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Summary. A materials research program is described. The program objectives are to evaluate materials and develop processing and fabrication techniques which it will be economically feasible to use in the construction of a high-gradient (10 MeV/ft) two-mile linear accelerator. RF properties are evaluated by cavity Q measurements at selected frequencies in the range 3 to 11 GHz. Techniques for reducing field emission are being sought. At present, this phenomenon is being studied by making dc measurements at room temperature. RF superconducting properties are known to be very sensitive functions of material purity and crystal structure. For this reason, various methods of deposition, surface purification and heat treatment are being tried. Most work is being done with lead and niobium, but technetium plating is being actively investigated.

Introduction

The very low RF surface resistance of superconductors together with the superfluid properties of liquid helium below the lambda point make high duty cycle high gradient linear accelerators an attractive possibility. Measured properties of superconductors indicate that materials exist which could conceivably provide gradients of 10 MeV/ft in a practical accelerator. On the basis of such measurements a feasibility study of a two-mile 100 GeV accelerator has been undertaken.[1] However, properties of many superconducting materials have been investigated only at dc or at very low frequencies and the preparation of surfaces of materials to operate at microwave frequencies with gradients of 10 MeV/ft is beyond the present "state-of-the-art." The present limitations are discussed here together with description of work in progress to achieve very high gradients in superconducting RF cavities suitable for use in linear accelerators.

Limitations

The first requirement of a material is that it will give a sufficiently low value of the surface impedance at the operating temperatures and frequencies. The RF surface impedance of an ideal superconductor decreases rapidly below the critical temperature, approaching zero at the absolute zero of temperature. However the imperfect nature of material surfaces gives rise to a residual resistance. These residual effects may be due to impurities in the surface, crystal grain boundary defects, a layer of oxide or condensate on the surface leading to dielectric loss, etc. In fact we lump into the residual resistance our ignorance of the exact nature of the material surface. By definition the residual resistance is obtained for vanishingly small stored energy in the cavity i.e., it is not dependent on signal level. A first objective in preparing cavities is to consistently obtain low-enough residual resistances (i.e., high Q improvement factors), to make the necessary refrigeration economically feasible. The surfaces must eventually be prepared in a manner which, on a large scale, is economically feasible.

After high Q-improvement factors have been obtained in cavities, their behavior at high field strengths must be considered. The high field behavior of superconducting cavities can be divided into two categories:

1. Effects due to the presence of high RF magnetic fields at the surface.
2. Effects due to high values of RF electric fields.

Materials for superconducting accelerators are operated in the region of perfect diamagnetism; that is, at magnetic fields below the level at which flux tube penetration takes place. Penetration of flux tubes with concomitant Q degradation is the problem met at high magnetic fields. High electric field effects are of at least three types; these are field-emission loading, electrical breakdown in the vacuum, and nonlinear dielectric losses.[2]

Magnetic Field Degradation

The superconducting state is destroyed when an applied dc magnetic field reaches a critical value, $H_c(t)$. At the critical temperature T_c, above which the material is normal, $H_c(T_c)$ is zero, while it increases to its largest value at $T = 0^\circ$K. The microwave critical magnetic field has been demonstrated to agree with dc measurements only for the type I superconductor tin.[3] These measurements were performed at fields below 200 gauss. On other materials fabricated into microwave cavities, Q degradation effects have occurred at magnetic fields less than the dc values. For the type II material, Nb, large decreases in Q have been observed at definite values of RF magnetic field well below the dc values. A sharp transition to the normal state is not seen, and increased surface purity, smoothness, and grain size lead to increased values of apparent $H_c(T)$. The type II elements, niobium and technetium, have very high values of first critical magnetic field $H_{c1}(T)$, measured at low frequencies, but these values have not been reached in RF measurements to date.

Electric Field Degradation

Large electric fields at the walls of the cavities can cause appreciable field emission currents. These field-emitted electrons are accelerated in the fields of the cavities causing power dissipation on the cavity walls which leads to unacceptable loading of the refrigerators.

Even though an electric field of 10 MV/ft = 3.3×10^5 V/cm is well below the threshold for measurable field emission, whiskers (microprotrusions) can easily enhance the electric field locally at their tips by a factor of more than 200. With a microscopic electric field of 6.6×10^7 V/cm, field emission current densities $\sim 10^7$ to 10^8 A/cm^2 may be obtained.[4]

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The average field emission power dissipation per emitter in the case of sinusoidal RF fields is:[5]

$$P = \frac{K d \beta^{\frac{5}{2}} E_0^{\frac{7}{2}}}{2\pi c} e^{-c/\beta E_0} \qquad (1)$$

where E_0 is the macroscopic accelerating electric field, d is the distance the field-emitted electrons have been accelerated when they impact, β is the enhancement factor, and K and c may be treated as constants. In the case of an accelerating field of 10 MV/ft, a work function of 4.7 eV, d = 3 cm, and $\beta = 200$, the power loss is ~10 W/emitter. If the enhancement factor were reduced by only a factor of 3, the power loss would be dramatically reduced to $\sim 10^{-10}$ W/emitter.

A power dissipation exceeding 10^{-1} W/ft can be a serious matter in a superconducting accelerator, due to the low thermodynamic efficiency of the refrigeration process at very low temperatures. Therefore, it should be clear that unless something is done to reduce and maintain low field enhancement factors throughout the accelerator structure, the field emission power loss will not be negligible — and may even prevent operation of the accelerator at high fields.

One method for reducing whisker size is known as selective sputtering, in which only the whiskers are sputtered off — the rest of the surface remaining essentially intact. The procedure is to introduce an inert gas such as He, while the whiskers are field-emitting with a current density $\sim 10^7$ A/cm^2. One theory is that ionization of the gas occurs in the region of high current density immediately in front of the whisker. The ions produced then fall in directly on the whisker due to the focusing action of the high electric field. Although there is not unanimity on the precise model to account for the result,[6] this method is quite effective in reducing the field enhancement.

With Pb deposited on Cu, it has been observed at SLAC that at a moderate dc emission current of 1×10^{-8} A, and argon pressures between 4×10^{-6} torr and 1×10^{-4} torr, β changes quasi-periodically. The length of the period appears to increase with increasing pressure. The enhancement factor both decreases and increases relative to its initial value, even after two days of sputtering. One may stop the sputtering with β either in a state of decrease or increase, but at these pressures and currents, the decrease is only minor—the emission current decreasing at most by a factor of 3. With an increased emission current of 1×10^{-6} A, and P = 1.4 $\times 10^{-4}$ torr, a permanent significant decrease in β was obtained. Within a few hours, the emission current showed a monotonic decrease which persisted over 24 h. The emission current was decreased by a factor of 140 (from 1×10^{-6} A to 0.7×10^{-8}A), and β was permanently reduced from 350 to 170. The experiment was not optimized, so that even greater reductions may be expected. Experiments are being conducted with helium which indicate that it is also effective in reducing β. Since argon and all the other inert gases would be solid below the superconducting transition temperature, it is important to establish that helium could be used for selective whisker sputtering.

A substantial decrease in field-emitted current can also be achieved by coating the metallic surface with a dielectric. This can reduce the field-emitted current by as much as 4 orders of magnitude.[7] An ideal dielectric coating would suppress field emission, increase breakdown voltage, contribute only a small dielectric loss, and be self-healing in case of electrical breakdown. We are investigating Al_2O_3 coatings and condensed gases. A coating of condensed gas such as argon represents an interesting possibility as this would take advantage of the self-healing properties of condensed gases in a low temperature environment. If, as suggested in Ref. (8), the dominant dielectric loss mechanism for condensed noble gases is radiation damping, a rough calculation shows that at 3 GHz, $\tan\delta \sim 10^{-25}$ for He or Ar, resulting in negligible loss.

Environmental Conditions

The high Q and the high field gradient properties of superconducting cavities must be maintained under all environments which can be reasonably expected during normal operation of a superconducting accelerator. The most important are repeated thermal cycling, accidental admission of wet air and radiation. We now consider these in some more detail.

Thermal Cycling

A bake-out of an accelerator structure is normally necessary and, depending on the material requirements, might require temperatures above 1000°C. The structure subsequently must be cooled to about minus 271°C. Over this range of temperature metallurgical changes take place in the material and these can affect the superconducting properties. These effects are particularly important when a layer of superconducting material is deposited on another substrate; for example, niobium or lead on copper. It appears that differential expansion between the deposited layers and the substrate is particularly injurious to the superconducting properties. Lead-plated copper cavities cycled from 70°C to 2°K at SLAC grow many large protrusions (the biggest ~500 μm long × ~1.6 μm diameter) of lead from the lead surface, usually at copper grain boundaries. At low gradients these protrusions have a small though significant effect on the Q of the cavities. They are expected to have a quite serious effect due to field emission losses when the cavity is subjected to high electric field gradients.

Leaks

It is prudent to also consider the effects of accidental atmospheric air leaks into the accelerator structure. Lead has been shown to be troublesome in this regard and shows oxidation on such exposure. On the other hand, in experiments with small X-band cavities, niobium recovers completely after being exposed to the atmosphere.

Radiation

Since the superconducting properties of a material are extremely structure-sensitive, it is very important to consider the effect of lattice defects which may be caused by irradiation during accelerator operation. This is especially true for niobium and technetium, which are type II materials. Experiments have shown that fast neutron radiation does change the K value of type II material, at high dose rates. Magnetization curves on niobium have been experimentally observed at Oak Ridge[9] to approach the behavior of a completely reversible type II superconductor, after radiation. From the accelerator standpoint, the radiation environment in these experiments was extreme, but radiation damage must be considered in the development of suitable superconducting materials.

Experimental Program

The program objectives are to evaluate materials and develop processing and fabrication techniques which it will be economically feasible to use in the construction of a high-gradient two-mile linear accelerator. Materials preparation techniques are studied and the corresponding RF superconducting properties are evaluated. The materials currently under study are lead, niobium and technetium.

As discussed above, electro-plated lead surfaces on copper do not respond well to thermal cycling or exposure to wet air. Niobium, which has a large dc value of H_{c1} and gives an adequate cavity-Q improvement factor, is being investigated. Surfaces prepared both by machining from solid stock and by molecular deposition on substrates are being tested. The methods of preparation are sputtering on substrates at elevated temperature, chemical vapor deposition on substrates, and machining or forming from solid material.

The radioactive (5×10^5 year half-life) element technetium is worth consideration as a practical superconductor. While the values of critical temperature and H_{c1}[10] are intermediate between lead and niobium, it has the advantages over lead that it is a hard, oxidation resistant metal, and the advantage over niobium that it can be electro-plated from an aqueous solution at room temperature.[11] For these reasons, an attempt is being made to build and test a technetium-plated copper cavity. The technique for electro-plating technetium on copper is being explored and very smooth surfaces on a microscopic level have been obtained. Some difficulty with surface cracking is being encountered at the present but it is expected that this will be avoided by proper adjustment of the electro-plating parameters. Plating is carried out completely in a vented glove box by a method developed at Oak Ridge National Laboratories.[11] No extraordinary difficulties have arisen.

Standard metallurgical techniques are being employed to examine the properties of the surfaces. Optical and Auger electron spectrometry methods will be used to detect trace impurities affecting the superconducting properties.

Surfaces are evaluated for their breakdown and field emission properties at dc levels in the apparatus sketched in Fig. 1. Provision is made to vary the separation between the two electrodes and to establish high electric fields by applying voltages up to 150 kV. An ultra high vacuum is maintained in the apparatus.

Evaluation of the electromagnetic properties of materials has been carried out at low frequencies and at microwave frequencies. Since very small samples may be conveniently tested at low frequencies, such measurements serve as a preliminary evaluation mechanism. Tests are done on samples in the form of rods a few inches long and less than 1/8 of an inch in diameter. A schematic of the measurement equipment is shown in Fig. 2. Signals from the secondaries of two pairs of coils are balanced, and a null is established on a very sensitive phase detector. The magnetic field is increased from zero to some high value and the change in susceptibility of the sample, which is the central core of one of the pairs of coils, is observed as a signal from the phase-locked detector. The behavior of an annealed sample of niobium is shown in Fig. 3. The departure from the perfect diamagnetic state at the critical magnetic field is observed as a sharp unbalancing of the transmitted signals. This apparatus is being utilized to observe the effect on the critical magnetic fields of various processing techniques and environmental conditions. This approach enables many small samples of materials to be investigated in a short time. Furthermore, it is hoped to achieve a sensitivity in the experiment which makes it possible to detect the penetration of a few flux tubes — which would be enough to reduce the Q value at microwave frequencies.

Surfaces which show promise then proceed to tests at microwave frequencies. These tests take place in magnetically shielded liquid helium dewars with highly stable tunable RF sources at selected bands of frequencies in the range 3 to 11 GHz.

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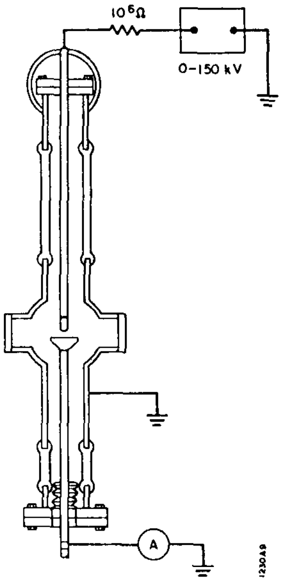

FIG. 1--Diagram of dc field emission system

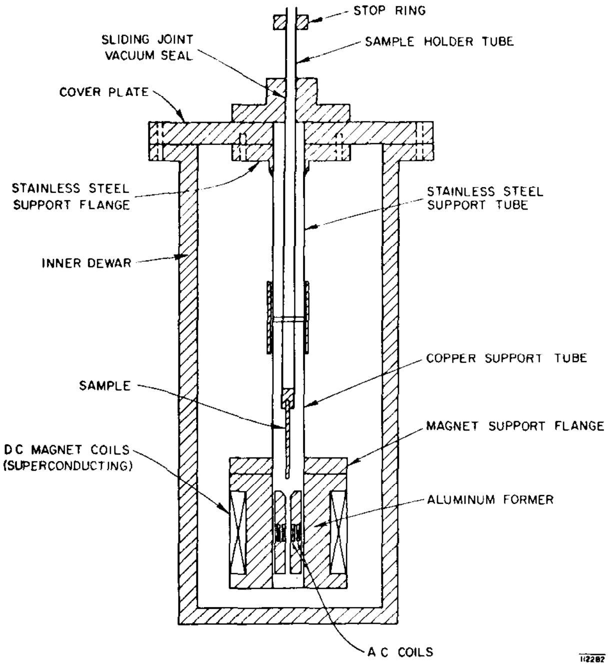

FIG. 2--Magnetic susceptibility measuring equipment

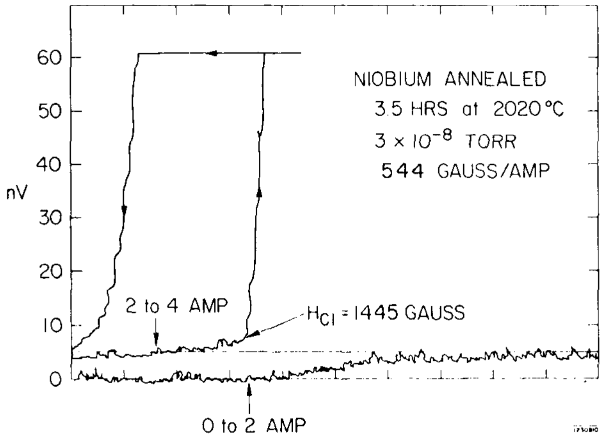

FIG. 3--Coil unbalance as a function of applied magnetic field for niobium sample