Progress Report No. 10

on

Thermal and Electrical Measurements on

Solids at Low Temperatures

submitted by

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This report summarizes the work done on this project from May, 1975 to the present. The participating staff consisted of H. Weinstock (Professor of Physics), P. J. Silverman (Assistant Professor of Physics), S. L. Colucci, A. R. Moodenbaugh and Y. L. Pak (Research Associates), the following graduate students: J. L. Ho and C. W. Thompson, Jr., and undergraduate students: R. Osa, W. Wild and P. Yarnold.

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I. SUMMARY OF RESEARCH PROGRESS

A. Effects of Low Temperature Fast Neutron Irradiation on Nb₃Sn Composite Wires in Fields Up To 10 Tesla

Determining the effects of neutron damage on the superconducting properties of Nb₃Sn is of importance in the design of magnets for controlled thermonuclear reactors. To approximate the reactor magnet environment, superconducting wires of Nb₃Sn were irradiated at low temperatures with fast neutrons. The induced changes in critical current density, j_c , were measured in a larger magnetic field than has been available at other facilities. From a basic point of view, these measurements provide insight into flux pinning mechanisms and their behavior.

Two types of Nb₃Sn composite wire were prepared by Dr. M. Suenaga of Brookhaven National Laboratory using the surface reaction of Nb wire codrawn in a Cu-Sn matrix. Both types were reacted at 700°C for 24 hours. One type (hereinafter referred to as type A) contained 19 Nb₃Sn filaments with a total cross sectional area of $7.4 \times 10^{-6} \text{ cm}^2$ and had a pre-irradiation $j_c = 1.0 \times 10^6 \text{ A/cm}^2$ at $H = 4 \text{ Tesla}$.¹ The improved type (hereinafter referred to as type B) contained but one Nb₃Sn filament of $5.7 \times 10^{-6} \text{ cm}^2$ cross section with a pre-irradiation $j_c = 1.6 \times 10^6 \text{ A/cm}^2$ at $H = 4 \text{ T}$.

Neutron irradiation was carried out at a temperature of 6 K in the fast neutron cryogenic facility of the CP-5 reactor at Argonne National Laboratory². A uranium converter and a concentric boron carbide cylinder surrounding the cryostat produce a neutron flux on the order of 10^{12} n/sec with essentially a fission spectrum ($E > 0.1 \text{ MeV}$). The wires were cut into 5 cm lengths and placed into an open-topped cylinder which was then inserted into the reactor.

After irradiation the cryostat was warmed to $\sim 70 \text{ K}$ and filled with liquid nitrogen. It was then possible to transfer the cylinder containing

the samples into a portable liquid nitrogen dewar while always maintaining the liquid level above the samples. Just prior to measurement, the wires were mounted onto a sample holder and placed into a bore of a superconducting magnet without ever warming the sample about 77 K.

It has been observed that annealing low temperature neutron damaged Nb_3Sn to 77 K changes j_c by less than 20%.^{3,4} Recovery below 77 K is due to point defect migration, and the remaining damage is in the form of more stable defect cascades which are the predominant radiation induced flux pinning centers.⁴

Current was passed through the sample via indium coated copper clamps. Low pressure contacts measured the potential developed across a one centimeter central segment of the sample. Each sample was positioned within the 2 inch bore of a superconducting magnet capable of producing transverse fields (at 4.2 K) of 11.5 T. At a given field the voltage as a function of transport current, $V(I)$, was displayed on an X-Y recorder. The critical current, I_c , was defined as the net current through the sample when $V(I)$ equalled 27 V.

Potentially destructive heating of the wire after the transition to the normal state was prevented by installing a manganin shunt of negligible magnetoresistance near the sample. By calibrating $V(I)$ for the shunt with the sample removed, I_c could be deduced from the total parallel circuit current. The small fraction of total current flowing through the shunt (5-6%) served to indicate that good current lead contact was made with the sample.

Figure 1 shows the behavior of type A material with irradiation and annealing. At a fluence of $4.8 \times 10^{17} \text{ n/cm}^2$, there was an overall increase in $j_c(H)$ above $H \approx 1.5 \text{ T}$.⁴ The enhancement in j_c becomes larger with field,

Fig. 1. Material A - Critical current density as a function of transverse magnetic field for several radiation doses and annealings.

Fig. 2. Material A - Volume pinning force density as a function of transverse magnetic field computed for some cases shown in Fig. 1.

increasing to 45% at 10 T. Increasing the fluence to $1.1 \times 10^{18} \text{ n/cm}^2$, for a second group of type A wires also produced an enhancement in $j_c(H)$ for $H > 2 \text{ T}$, but the increase was not as great as in the lower dose samples. Although each curve represents measurements done on a different wire, data shown for two samples with the same higher dose demonstrate reproducibility.

Annealing the low dose sample to room temperature lowered the $j_c(H)$ toward that for an unirradiated specimen. At the largest fields of measurement, j_c for the annealed wire is quite close to that for an unirradiated wire. In vacuo annealing of the high dose sample to 415 K produced significant changes in $j_c(H)$. Below about 4 T the critical current was substantially below that for the unirradiated sample, and at about 10 T the two curves apparently converge.

The corresponding volume pinning force density ($\vec{F}_p = \vec{j}_c \times \vec{B}$, where $B \approx H$ for $H \gg H_{c1}$) is shown as a function of field in Fig. 2. For the unirradiated type A sample, $F_p(H)$ has a maximum of $4 \times 10^9 \text{ dyn/cm}^3$ occurring at about 2.75 T. $F_p(\text{max})$ increased 22% and shifted to 4.25 T after a $4.8 \times 10^{17} \text{ n/cm}^2$ irradiation. A larger dose of $1.1 \times 10^{18} \text{ n/cm}^2$ decreased the magnitude of $F_p(\text{max})$, but left the magnetic field value for $F_p(\text{max})$ unchanged. Annealing the low dose sample to room temperature lowered $F_p(H)$ and $H(F_p(\text{max}))$. Annealing a high dose sample to 415 K, lowered $F_p(H)$, but increased $H(F_p(\text{max}))$. This shift is related to the low field degradation of $j_c(H)$. The high field $F_p(H)$ values for irradiated wires seem to approach the same functional dependence, an effect observed in other pinning studies.^{5,6}

The critical current variation for type B material as a function of field is shown in Fig. 3. Although the unirradiated $j_c(H)$ values in material B ($j_c(4 \text{ T}) = 1.6 \times 10^6 \text{ A/cm}^2$) are larger than those for material A, irradiating to a larger fluence of $1.8 \times 10^{18} \text{ n/cm}^2$ produced an even greater fractional increase in j_c . At 10 T the j_c enhancement approached 60%. In vacuo annealing

Fig. 3. Material B - Critical current density as a function of transverse magnetic field for unirradiated, irradiated, and irradiated and annealed specimens.

Fig. 4. Material B - Volume pinning force density as a function of transverse magnetic field computed for the cases shown in Fig. 3.

to 540 K reduced the enhancement by roughly one half, indicating the apparent stability of the radiation induced modification. The low field degradation of $j_c(H)$ with irradiation is again probably due to a slight reduction in T_c .

The volume pinning force density as a function of field for type B wires is shown in Fig. 4 and is qualitatively similar to the behavior for type A material. Irradiation increases the magnitude of $F_p(\max)$ and shifts $H(F_p(\max))$ to higher field values. Annealing reduces $F_p(H)$ somewhat, but leaves the position of the peak relatively unchanged. The overall larger $F_p(H)$ values for material B corresponds to its larger $j_c(H)$ values.

The greater enhancement of the properties in material B with irradiation was not expected. Enhancement of $j_c(H)$ by irradiation has been frequently observed^{4,7-9} and is attributed to the introduction of radiation induced flux pinning centers. The critical current can be expected to increase with radiation dose until the potential wells associated with pinning centers begin to overlap, when subsequent irradiation reduces $j_c(H)$. This is because the fluxoids experience an effectively reduced potential barrier and, hence, a decreased resistance to flux flow. It follows that materials in which pinning centers are initially optimized, show a degradation in $j_c(H)$ for even small radiation doses.¹⁰

In some superconducting materials, optimum $j_c(H)$ values are approached because it is possible to manipulate the number and size of grain boundaries, second phase precipitates, or other defects which form pinning centers. Since most flux pinning occurs at grain boundaries for Nb_3Sn ,¹¹ it is probable that $j_c(H)$ values are not now near optimization. This is because the combined temperature dependence of the Nb_3Sn growth rate with the grain size results in a relatively large grain.¹¹ As appears likely in this work, low temperature fast neutron irradiation introduces displacement cascades which act as

additional pinning centers to enhance $j_c(H)$, even for Nb_3Sn with large unirradiated $j_c(H)$ values.

It might be expected that the $j_c(H)$ enhancement with radiation would be less in the improved material B than in material A. The fact that just the opposite is true, has provided greater impetus to the current radiation damage study on Nb_3Sn . A comparison of the enhancements in type A and type B materials is shown in Fig. 5.

One objective to be pursued in conjunction with M. Suenaga is to determine how microstructure differences in the two materials might cause the observed behavior. Another objective is to perform additional irradiations and measurements in an attempt to better characterize this behavior. In summary, however, the large enhancement in $j_c(H)$ is most promising in regard to fusion reactor applications.

Fig. 5. Comparison of radiation-induced $j_c(H)$ enhancements in materials A and B.

B. Thermal Conductivity of Radiation Damaged and Deformed MgO

Due to a change in laboratory personnel, this effort was temporarily curtailed during the early part of the current academic year. However, this study now has been resumed. Recently, measurements have been carried out on a specimen with approximately a 2% deformation along a [100] direction, and a subsequent annealing treatment at 750°C. These measurements are an extension to lower temperatures of data obtained previously¹², but with anomalously large scatter. It now appears that the data follow approximately a T^3 thermal conductivity dependence in the temperature range of 1 K to 4 K and with a magnitude approximating that found for an undeformed, unirradiated specimen. This preliminary result tends to indicate that dislocation strain fields are not sufficiently abundant to produce a lower thermal conductivity, as seemed possible from the higher temperature data taken earlier and showing what might have been the onset of a $K \propto T^2$ dependence. Additional measurements on this annealed specimen are in progress.

During the current year discussion of the detailed study of defects in MgO (which have been carried out by this laboratory) has led to the formulation of plans to undertake a sequence of such studies using fast neutrons on potentially important insulating materials for nuclear fusion reactors. This is to be a cooperative effort carried out in conjunction with a group headed by Dr. Frank Clinard at the Los Alamos Scientific Laboratory, and utilizing irradiation by 14 Mev neutrons at the Lawrence Livermore Laboratory and by a fission neutron spectrum ($E > 0.1$ Mev) at the CP-5 reactor at the Argonne National Laboratory. During the current year arrangements have been made for this effort through frequent communications between Dr. Clinard's group and this laboratory, and through independent study. A fuller discussion of plans is presented in the May 1976 Renewal Proposal.

C. Thermal Conductivity of Neutron Irradiated Superconducting Vanadium

Preliminary measurements - see Progress Report No. 9 - on two high purity single crystal specimens of vanadium obtained from Dr. S. T. Sekula of Oak Ridge National Laboratory have been suspended. This was done when it was found possible to obtain an even higher purity specimen through the courtesy of Professor G. Danielson and his collaborators at the ERDA Ames Laboratory. This specimen has a room temperature to liquid helium temperature electrical resistance ratio of 1740, as compared to a 630 ratio for the two specimens from Dr. Sekula.

Currently, measurements of the thermal conductivity of this new sample are in progress, and will be compared to the earlier obtained data for both the superconducting and normal states of the less pure samples mentioned above. When this has been completed, a sequence of neutron irradiations will be carried out at ANL's CP-5 reactor, with low temperature thermal conductivity data to be obtained between each reactor exposure. The present measurements had been held up by the demise of the original laboratory superconducting magnet (about 10 years old), which is the only one in the laboratory suitable for use with the thermal conductivity cryostats, and which is required to operate in the persistent mode when normal state data are being taken. A replacement magnet is currently under construction and is expected to be ready for use within two to three weeks. Measurements in the superconducting state are now being made.

D. Other Thermal Measurements, Construction of Facilities and Crystal Growth

A continuation of thermal conductivity measurements on tantalum wires used in conjunction with the analysis of normal-superconducting interface velocity data (obtained at the Los Alamos Scientific Laboratory by Dr. W. C. Overton, Jr. and the Principal Investigator) was attempted. Unfortunately, measurements for the normal state conductivity were thwarted by the malfunctioning of the superconducting magnet, a fact alluded to in the previous section. Despite a number of attempts to repair the magnet locally, it was found that it could not function in the persistent mode at all, and in an externally driven current mode it was found to create too much noise to allow for reliable thermal conductivity data. Interim plans (while a new magnet was being requested) had been made to utilize a larger magnet and dewar system which could accommodate the thermal conductivity cryostat, albeit with a much larger expenditure of liquid helium and liquid nitrogen. However, Dr. Pak who was carrying out this study resigned in mid-year to accept a position in industry which he was forced to decide upon with only a limited time available.

In view of the cited problems with the superconducting magnet used in several thermal conductivity studies, and the unprecedented turnover in laboratory personnel during the past year - only one research associate (with one year's experience) and the Principal Investigator are "holdovers" - it was decided to make a major effort to overhaul some well-worn experimental cryostats, to build a metal liquid helium-liquid nitrogen dewar set which would put less dependence on those currently in use, and to complete the construction of a helium dilution refrigerator for which most major components were already on hand. This "building program" has had the effect not only

of upgrading and expanding laboratory facilities, but has had (and is continuing to have) the effect of educating the new laboratory personnel. These include an assistant professor, a post-doctoral research associate, two graduate students and two undergraduate students.

A very small scale program in growing from solution a variety of anti-ferromagnetic insulating crystals has been continued.

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12. J. B. Hartmann, H. Weinstock and Y. Chen, in Phonon Scattering in Solids, edited by L. J. Challis, V. W. Rampton and A. F. G. Wyatt (Plenum Press, 1976, New York) p. 46.

II. SUMMARY OF PUBLICATIONS: May 1975 to the present

1. "Thermal Conductivity of Radiation Damaged and Deformed MgO";
J. B. Hartmann, H. Weinstock and Y. Chen
MgO Workshop, Canton NC, May 26-28, 1975; (COO-1629-49)
2. "Heat Conduction in Superconducting Lead Thallium Alloys"
J. L. Ho
Ph.D. Thesis, IIT (December 1975); (COO-1629-54)
3. "Effects of Low Temperature Neutron Irradiation on Nb₃Sn Composite Wires
in Fields Up to 10 Tesla";
S. L. Colucci, H. Weinstock, B. S. Brown, and T. H. Blewitt
Low Temperature Physics - LT14, edited by M. Krusius and M. Vuorio
(North-Holland, 1975, Amsterdam) vol. 2, pp. 9-12; (COO-1629-50)
4. "Phonon Thermal Conductivity of Superconducting Lead-Thallium Alloys";
J. L. Ho, C. K. Chau, H. Weinstock, and W. C. Overton, Jr.
Low Temperature Physics - LT14, edited by M. Krusius and M. Vuorio
(North-Holland, 1975, Amsterdam) vol. 2, pp. 9-12; (COO-1629-51)
5. "Thermal Conductivity of Radiation Damaged, Additively Colored and
Deformed MgO";
J. B. Hartmann, H. Weinstock and Y. Chen
Phonon Scattering in Solids, edited by L. J. Challis, V. W. Rampton, and
A. F. G. Wyatt (Plenum Press, 1976, New York) pp. 190-192; (COO-1629-52)
6. "Critical Currents in Nb₃Sn Fast-Neutron Irradiated at 6K";
S. L. Colucci, H. Weinstock and B. S. Brown
Applied Physics Letters 28, 667 (1976); (COO-1629-55)
7. "Critical Currents in Nb₃Sn Fast Neutron Irradiated at 6K";
S. L. Colucci and H. Weinstock
Bull. Am. Phys. Soc. 21, 289 (1976); (COO-1629-56)

III. GRADUATE STUDENT PROGRESS

Jen-Lie Ho was awarded the Ph.D. degree in August, 1975 and immediately began full-time employment with the Burroughs Corporation.

Charles W. Thompson, Jr., a graduate student who has completed all but the thesis related requirements for the Ph.D. degree, has joined the laboratory during the current academic year. Mr. Thompson, holder of a B.A. from Amherst College and an M.A. from the University of Chicago, has been awarded a \$3,600 fellowship for one year beginning July 1, 1976. He was chosen by the Selection Committee for the 1976-77 Graduate Fellowships for Black Americans of the National Fellowships Fund (located in Atlanta, GA). His thesis study will involve thermal conductivity measurements on a series of superconducting Pb-Bi alloys now being prepared at the Los Alamos Scientific Laboratory. The aim of this work is to study electron-phonon coupling.

Donald Samuels, a first year graduate student currently enrolled as a candidate for the M.S. degree, has agreed to participate in the laboratory program actively beginning in September. For the interim period he is engaging in a reading program supervised by the Principal Investigator. His M.S. thesis problem will probably involve thermal conductivity measurements on annealed and strained specimens of superconducting Pb-Tl alloys used earlier in the Ph.D. thesis research of Dr. Ho. If Mr. Samuels successfully passes the Ph.D. qualifying exam being offered in January 1977, he will be given a more advanced problem after conclusion of the Pb-Tl study now contemplated.

Richard Osa, an undergraduate student who has contributed significantly to the laboratory effort for almost three years, is being graduated this month with a B.S. degree in Physics. He has just accepted an offer of a \$5,000 Research Assistantship in the Materials Science Department of Northwestern University commencing September 1976.