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RADIATION EFFECTS LIMITS ON SUPERCONDUCTING
MAGNETS: DATA BASE FOR COPPER STABILIZERS

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3.4 RADIATION EFFECTS LIMITS ON SUPERCONDUCTING MAGNETS: DATA BASE FOR COPPER STABILIZERS — M. W. Guinan (Lawrence Livermore National Laboratory)

3.4.1 SPM Task

IV.G.2 - Radiation Damage in Superconducting Composites

3.4.2 Objective

The objective of this work is to quantify the changes which occur in the magnetoresistivity of coppers (having various purities and pretreatments) at fields up to 12T during the course of sequential neutron irradiations at 4K and anneals to room temperature. In conjunction with work by Klabunde and Colman at ORNL, the results should lead to engineering design data for the stabilizers of superconducting magnets in fusion reactors. These magnets are expected to be irradiated during reactor operation and warmed to room temperature periodically during maintenance.

3.4.3 Summary

Two of eight differently prepared copper stabilizer samples, previously irradiated in the RTNS-II at LLNL, the IPHS-I at ANL, and the BSR at ORNL, have been irradiated to a fluence of 1.33×10^{22} n/m² at RTNS-II. During the course of the irradiation the samples were periodically removed (without warming) for measurements of the transverse magnetoresistance and returned for continued irradiation. This experiment extends the range of neutron irradiation induced resistivity by a factor of five over the previous experiments. A simple model is developed which reproduces the magnetoresistance results of all the experiments to an accuracy of 2.5%.

3.5.4 Progress and Status

3.5.4.1 Introduction

In order to predict the response of a large superconducting magnet to cyclic irradiation and annealing, the magnet designer requires four pieces of information:

1. A detailed knowledge of the neutron spectrum in the magnet.
2. The rate of resistivity increase at zero field in this spectrum.
3. The dependence of annealing at room temperature on initial purity, cold-work and neutron spectrum and fluence.
4. The resistivity at field arising from variations in the zero field resistivity due to initial purity, irradiation, cold-work and annealing.

The importance of item one cannot be over-emphasized. It is potentially the largest source of error in predicting magnet response.

Given the adequacy of item one, the initial damage rate can be predicted from damage energy calculations and a knowledge of the damage efficiency as a function of recoil energy.^{1,2} At present, an accuracy of 10% or better is achievable. Tables of initial damage rates as a function of neutron energy are available.¹

The annealing response is sensitive, i.e. up to a factor of two, to initial purity, cold-work and neutron spectrum and fluence.^{1,3} We will present only a rough correlation of the available data. Further work is required at higher fluences.

The main concern of this report will be directed at correlating all the data available from a previous RTNS-II experiment,^{1,3} the experiment at the ANL/IPHS-I Facility by Klabunde and Colman⁴ and the present higher dose results.

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3.5.4.2 Samples

The details of sample preparation are given in previous reports.³⁻⁵ Two samples, one of 99.999% recrystallized copper from ASARCO with an oxidizing anneal, and a second of OFHC copper from Intermagnetics General Corp., unoxidized, with 7.5% cold-work were utilized in the latest experiment.⁵ The first, designated ASARCO-0, had a residual resistivity ratio ($R_{293}/R_{4.2}$) of 1780. The second, designated OFHCUN-1, had a ratio of 166. Both had 10 mm gauge lengths.

3.5.4.3 Experimental Procedure

The ASARCO-0 and OFHCUN-1 samples were irradiated in five segments to final fluences of 1.20×10^{22} n/cm² and 1.33×10^{22} n/m², respectively. At the end of each segment, the magnetoresistance was measured at 2, 4, 6, 9, 10 and 12T. During three of the transfer operations to the 12T magnet, unintentional temperature excursions to 40-50K were experienced. Complete details of the experiment are given in Ref. 5.

3.5.4.4 Results and Discussion

From the analysis of the raw resistance data, resistivities were determined for each sample assuming a thermal resistivity of $17.86 \mu\Omega\text{-m-m}$ at 35.1°C. All values were then corrected for electrical size effects.³ This is the same procedure followed in the other experiments.³⁻⁵

Annealing data

In order to compare the annealing data in our previous work^{1,2} with that at IPNS-1⁴ and BSR⁵, we needed a method of data correlation to compare experiments with different resistivity increments per cycle. An obvious choice was to plot the accumulated annealed resistivity ($\Delta\rho_{ANN}$) vs. the accumulated resistivity at 4.2K ($\Delta\rho_{TOT}$). $\Delta\rho_{TOT}$ serves the purpose of an exposure parameter and in fact is proportional to neutron fluence at low exposures. Furthermore, it allows us to directly compare experiments in different environments. For the purpose of estimates of retained damage, we have plotted $\Delta\rho_{ANN}/\Delta\rho_{TOT} \times 100$ (i.e. % of total damage retained) as a function of $\Delta\rho_{TOT}$ for three cases. The first, Fig. 3.5.1, shows the data for sample OFHCUN-1 in three spectra. The RTNS data represent a spectrum much harder than that generally expected, the BSR data a much softer spectrum, and the IPNS data one comparable to magnet spectra. As the figure shows, the retained damage varies by a factor of two, with more remaining in the harder spectra.

Fig. 3.5.1 The dependence of the retained resistivity ($\Delta\rho_{ANN}/\Delta\rho_{TOT}$) on the accumulated resistivity at 4K ($\Delta\rho_{TOT}$) for three neutron spectra; RTNS-11(Δ), IPNS-1($\diamond$), and BSR($\square$) for sample OFHCUN-1.

Fig. 3.5.2 shows the effect of cold-work on retained damage. The RTNS-II irradiation data are for samples OFHCUN-0, 1 and 2 with 0%, 7.5% and 14.3% cold-work respectively. As expected, the difference due to cold-work diminishes as the exposure is increased. This is the result of the buildup of annealed damage and irradiation modification of the microstructure. At very high exposure we expect the samples to approach a single microstructure.

The results for the IPHS-I irradiations are qualitatively similar, since these are at lower damage levels, the differences due to cold-work are larger. At the only overlapping exposure (about 30×10^{11} T/n-m) corresponding to our first cycle, the retained defect resistivity varies from 0.7 to 0.8 of ours with the lower values corresponding to the largest amount of cold-work.

The influence of initial purity is shown in Fig. 3.5.3, where results for recrystallized samples ASARCO-0, OFHCUN-0 and OFHCQX-0 are shown. Comparing these results with those in Fig. 3.5.2, we see that an increase in purity corresponding to a reduction 2×10^{11} T/n-m has about the same effect as increased cold-work corresponding to an increase of 5×10^{11} T/n-m. Again the results are qualitatively similar to that observed at IPHS-I. Differences in the retained defect resistivity are almost independent of dose. At 30×10^{11} T/n-m exposure, the IPHS-I retained resistivities were about .80 of those at RTNS-II.

The shape of curves in Figs. 3.5.1, 2 and 3 suggest a reciprocal damage rate model², developed for electron irradiations, i.e. $\Delta\rho/\Delta\phi$ vs. $\Delta\phi$ might provide a linear fit to the data. In fact such a fit has been attempted to room temperature irradiated copper in both a reactor³ and RTNS-II.¹⁹ All the RTNS-II plots show a slight but real upward curvature, while the IPHS plots show both positive and negative curvatures. The resulting latitude in slope and intercept is sufficient to preclude any simple interpretation in terms of impurity or cold-work effects. The results are close enough to linear, however, that an attempt to develop a similar model in terms suitable for cascade defect production should prove fruitful.

Magnetoresistivity

In our earlier work^{1,2} at RTNS-II we reported that the effects of irradiation, cold-work and annealing on a given material (OFHCUN) all fell on a single Kohler plot, i.e.

$$\rho(H) = \rho(0) / \rho(0) = f(H/\rho(0)). \quad (1)$$

This appeared to be true over the limited range of $H/\rho(0)$ for the OFHCUN samples, 0.1 to 2×10^{11} T/n-m. When the higher dose data and the results from IPHS-I became available, it was clear that this was no longer a valid conclusion, particularly at low values of $H/\rho(0)$.

The present data set, from all three experiments, cover the range of $H/\rho(0)$ from 0.015 to 12.5. When all of the data are plotted on a single Kohler plot, a pronounced fan develops in the region below about 0.5×10^{11} T/n-m. At 0.015 the spread amounts to a factor of 4 in $f(H/\rho(0))$. While this amounts to an uncertainty in $\rho(10T)$ of only 12-15%, after 5-10 anneal cycles the lifetime could be changed by up to 50%. To try and account for the spread, we begin eliminating any data set which produced points in the fan central region. At the end of this process only two branches were left. The remaining data are shown in Fig. 3.5.4. The sets of files giving rise to the two branches were distinguishable. One set, the lower, consisted of files for which 95% or more of the resistivity at zero field was the result of point defects or small clusters of point defects, i.e. the files for unirradiated, recrystallized samples and those for which no room temperature anneals had been carried out after irradiation. The remaining set consisted of files for which more than 90% of the resistivity arose either from cold-work and/or annealed radiation damage.

Since our final aim is only to develop an equation which will describe all the magnetoresistance effects arising from any combination of extended and point defects, any form will suffice. It would be better if the limiting behavior of $f(H/\rho(0))$ followed that observed; proportional to H at high fields and vanishing as H^2 at low fields.

Ziman¹¹ proposed a simple function for the conductivity of a polycrystalline sample with a multiply connected Fermi surface based on behavior expected for a simple model cylindrical Fermi surface averaged over all crystalline directions. In this case the electrical conductivity is given by

$$\sigma(H) = \sigma(0) (1/\omega_H \tau) \operatorname{atan}(\omega_H \tau) \quad (2)$$

where ω_H is the cyclotron frequency with the field parallel to the cylinder axis and τ is the electron collision time. Since $\omega_H = (e/m^*c)H$ and τ is proportional to $\sigma(0)$, we can rewrite eq. 2 as

$$\sigma(H) = (1/CH) \operatorname{atan}(CH/\rho_0) \quad (3)$$

where c is a function only of the density of states at the Fermi surface. In order to fit the data of Fig. 3.5.4, an initial attempt was made to fit the point defect data set to two such terms, reflecting the contribution of two sets of electrons with different Fermi surface orbits to the overall conductivity. This requires three parameters and provided only a poor ($\pm 12\%$) fit to the data.

Fig. 3.5.2 The dependence of the retained resistivity ($\Delta\rho_{ANN}/\Delta\rho_{TOT}$) on the accumulated resistivity at $4K(\Delta\rho_{TOT})$ for three levels of cold-work; OFHCUN-0(0%), OFHCUN-1(7.5%) and OFHCUN-2(14.3%).

Fig. 3.5.3 The dependence of the retained resistivity ($\Delta\rho_{ANN}/\Delta\rho_{TOT}$) on the accumulated resistivity at $4K(\Delta\rho_{TOT})$ for three levels of purity; ASARCO-0, OFHCOX-0 and OFHCUN-0.

Fig. 3.5.4 Kohler plots for samples for which the resistivity at zero field is due entirely to either point defects(Δ) or extended defects($\circ$).

Three such terms produced an excellent fit to the point defect data sets but require five parameters. The resulting equation for $f(H/p(0)) \equiv f(x)$ is

$$f(x) = \frac{1}{(1/CX) \operatorname{atan}(f_1 CX) + (1/DX) \operatorname{atan}(f_2 DX) + (1/EX) \operatorname{atan}((1-f_1-f_2)/EX)}^{-1} \quad (4)$$

As before C , D , and E depend only on the density of states at the Fermi surface. While f_1 , f_2 are partial contributions to the total conductivity at zero field, and are dependent on the scattering behavior of the electrons comprising that group. The third term in the denominator insures that the sum of the individual contributions add up to the total conductivity.

The point defect data set is shown in Fig. 3.5.5 with the fitted function. The standard deviation of the 126 data points from the fit is $\pm 2.1\%$. Values for the constants are given below with their corresponding standard deviations.

$$\begin{aligned} C &= 9.683 \pm .080 \\ D &= 153.5 \pm 17.3 \\ E &= 15.37 \pm .48 \\ f_1 &= .7698 \pm .0091 \\ f_2 &= .2049 \pm .0100 \\ f_3 &= .0263 \end{aligned}$$

While five parameters were required we could find no other 4 parameter fit using completely arbitrary functions, that did as well. We are now left to fit the data sets in which the resistivity arose entirely from extended defects. Since C , D , and E are material parameters, only f_1 and f_2 can vary. The resulting fit can be seen in Fig. 3.5.6. The additional 57 data points of this set are fit with a standard deviation of 2.9%. The value of f_1 decreased and f_2 increased as below, resulting in a net increase in f_3 .

$$\begin{aligned} f_1 &= .3436 \pm .0237 \\ f_2 &= .6266 \pm .0253 \\ f_3 &= .0398 \end{aligned}$$

We note that the resulting fit comes very close to the Kohler function found by Fickett¹² on high purity copper in which $\rho(0)$ was determined primarily by phonon contributions at low $H/\rho(0)$. We also note that within the accuracy of the fit we could hold the ratio of f_2 to f_1 constant implying that only the scattering of the f_1 electrons is changed by the introduction of extended defects.

To fit the remaining data, comprising an additional 1000 data points, we only need combine these two fits. Since the f 's are proportional to the partial conductivities, a fit to mixed point defect and extended defect sets would use values for f_1 and f_2 as follows

$$f_1 = f_{1PD} \rho_{PD}/\rho_0 + f_{1ED} \rho_{ED}/\rho_0 \quad (6)$$

where ρ_{PD} and ρ_{ED} are the portions of the resistivity at zero field arising from point and extended defects, respectively.

3.5.5 Conclusions

A simple model has been developed which can predict the magnetoresistance of copper as a function of initial purity, the extent of cold-work and the presence of both irradiation produced point defects and the extended defects produced by annealing. The remaining area of uncertainty in a complete description of the response of superconducting magnets to irradiation-anneal cycles is the dependence of annealing on dose, impurity concentration and cold-work.

3.5.6 Acknowledgment

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Fig. 3.5.5 The Kohler plot for samples for which the resistivity at zero field is due entirely to point defects. The solid line is the fit to the data (see text).

Fig 3.5.6 The Kohler plot for samples for which the resistivity at zero field is due entirely to extended defects. The solid line is the fit to the data (see text). The dashed line is the fit to the data of Fig. 3.5.5.

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