RADIATION EFFECTS ON SUPERCONDUCTIVITY

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Prepared For Presentation at

Materials Science Seminar
ASM

Cincinnati, Ohio

November 9 - 10, 1975
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December 1975


*Work supported by the U. S. Energy Research and Development Administration.
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INTRODUCTION

A discussion of the influence of radiation-induced defects in superconductors necessarily includes an understanding of the various properties of the superconducting state (1-3). The transition from the normal to the superconducting state is characterized by an abrupt drop to zero of electrical resistivity and also by a change in the material from nonmagnetic to perfectly diamagnetic. If an ideal superconductor is cooled below its superconducting transition temperature ($T_c$) in a magnetic field, where $H$ is less than a critical field, the field is excluded at $T = T_c$. The sample becomes fully diamagnetic as a result of lossless currents that circulate on the surface of the specimen. The exclusion of the field is complete for fields less than a critical field above which the field penetrates the sample. A type-I superconductor is characterized by complete penetration of the field above the critical field ($H_{c1}$). A type-II superconductor (Fig. 1) has complete magnetic field expulsion up to the lower critical field $H_{c1}$, but above $H_{c1}$ the magnetic field gradually penetrates the sample in regions of normal conducting filaments (flux lines or fluxoids) each carrying a quantum unit of magnetic flux ($\phi_0$) lying parallel to the imposed field. The flux within each fluxoid core is generated by a vortex of persistent current that circulates around the core, and the fluxoids repel each other. The field penetration is complete at the upper critical field $H_{c2}$ where the normal cores overlap and the remaining vestiges of superconductivity are destroyed. Since the main topic of the present review is the

*Work supported by the U. S. Energy Research and Development Administration.
interactions of fluxoids with various defects, only
type-II superconductors will be
discussed.

The flux lines
are arranged in a
periodic array, and
the flux-line lat-
tice (FLL) structure
depends on the field
orientation with re-
spect to the crys-
tal lattice. The FLL
has been directly observed by decorating the FLL in a thin-
foil superconductor with small ferromagnetic partic'es and
then inspecting a replica of the FLL arrangement in the foil
with transmission-electron microscopy (Fig. 2) (4). Simply
stated, the flux lines begin
penetration at $H_{c1}$ in the
mixed state, and their spac-
ing decreases as $H$ increases
until they overlap at $H_{c2}$. 
At $H_{c2}$, the field is fully
penetrating, and the sample
bulk is normal. In terms of
this concept, the spacing of
the flux lines in a tri-
angular lattice as a function
of field is $d_{FLL}$, where

$$d_{FLL} = \sqrt{1.16\phi_0 \over B}$$

$$= \frac{1520\, \mu}{\sqrt{B \text{ (in kG)}}} \quad (1)$$

Fig. 2. Cross-Sectional View
of a Replica of the Flux-Line
Lattice in a Lead-Indium Alloy. quantum, and $B$ is the magnetic
flux density. The diameter of
the fluxoid is roughly twice the coherence length $\xi$, a para-
meter that characterizes the distance over which the density
of superelectrons can change appreciably
diam = 2\sqrt{\frac{\alpha}{\beta}} = 2\sqrt{\frac{1128\text{ A}}{1.2\text{ kg}}} = 12.28 \text{ A}

The upper critical field is related to the thermodynamic critical field $H_C$ by

$$H_{c2} = v^2 \cdot H_C$$

where $v$ is the Ginzburg-Landau parameter. The Ginzburg-Landau theory for type-II superconductors shows that, near $T_c$, $v$ varies with the residual electrical resistivity, as (2)

$$v = v_0 + A \cdot \rho^{1/2}$$

where $v_0$ denotes the pure material, $A$ is a constant and $\rho$ is the electronic specific heat. Since $\rho$ is a sensitive function of the defect state of the crystal, radiation damage has a considerable effect on $v$ and therefore $H_{c2}$. The effect of damage on $H_C$ has been found to be large only in compound superconductors.

If no resistance to the motion of the flux lines occurs, the repulsion for $H = H_{c1}$ will result in a uniform distribution throughout the specimen and only the spacing of the flux lines will change with $H$. In real samples, flux lines not only interact with each other (giving rise to FLI elastic constants) but also with inhomogeneities and defects in the crystal. These interactions hinder the motion of flux lines with respect to the crystal lattice (flux pinning) and result in a nonuniform flux-line distribution. The "critical state" is characterized by an arrangement of flux lines such that local flux distributions result in a driving force equal to the pinning force of the defects.

When a transport current transverse to the applied field is applied to a superconductor in the mixed state, the fluxoids will feel a Lorentz force. The current will be transported in a lossless manner as long as the Lorentz force is less than the pinning force. The critical current density, $J_c$, is defined as the maximum current attained before the pinning forces are overcome, and, for high $\rho$ material,

$$F_p = |\vec{B} \times \vec{J}_c|$$

(5)
where \( F_p \) is the volume pinning force density. For \( J < J_c \), the fluxoids are un pinned and move through the crystal. This motion dissipates energy that must be supplied by the current source, and a voltage appears across the sample and the superconductor becomes resistive. Therefore, the critical current can be increased in materials by introducing defects of any type that will increase the volume pinning-force density \( F_p \). In the present review, \( F_p \) and \( J_c \) will be used interchangeably when discussing the flux-pinning experiments.

The aim of most radiation-damage experiments in superconductors is not only to increase \( J_c \) for technological purposes and understand the limitations in a radiation environment but also to understand the basic interaction mechanism between the flux lines and the various defects. Flux pinning in type-II superconductors results from crystalline imperfections that produce local fluctuations in the properties of a material (such as the electronic mean free path or the local phonon spectra) which make the free energy of the specimen depend on the position of the fluxoids. Significant pinning occurs only for pinning centers, the influence of which extends, at least in one direction, over a distance of the order of the coherence length \( \xi \), since the superconducting properties cannot change appreciably over distances less than \( \xi \). The state of understanding of the flux-pinning characteristics of various structures (such as dislocation loops, cell walls, voids, normal precipitates, and defect cascades) was contained in an excellent review by Campbell and Evetts (2) and was discussed at the International Discussion Meeting on Flux Pinning in Superconductors (3).

It is desired to relate the volume-pinning-force density \( F_p \) to the defect microstructure. The first step is to calculate the elementary pinning force \( f_p \), the maximum interaction force between a single pinning center and the flux-line lattice. It is then necessary to sum appropriately the individual \( f_p \)'s to obtain the volume-pinning-force density \( F_p \), which is related to the experimentally accessible \( J_c \). This summation is the difficult step because the flux lines interact with each other as well as with more than one defect. The complexity of this problem can be illustrated by discussing the analogous problem of mechanical hardening by a hard particle dispersion in a ductile metal. The hardening results from an interaction between dislocations and solute atoms. The dislocations are impeded by this interaction, and movement is possible only if the applied stress is greater than the critical shear stress. The problem is to calculate
both the interaction force between a dislocation and a single atom and the number of atoms contacted by the dislocation, which is a statistical problem.

It is also necessary, in the case of flux pinning, to calculate the number of pins contributing their $f_p$ to $F_p$. If the FLL had no rigidity (i.e., no flux line repulsion) the flux lines could arrange themselves so that the maximum number were interacting with the attractive pinning centers and the total pinning force would be a simple sum. The finite rigidity of the FLL (as expressed by the elastic constants $C_{eff}$) limits the number of pinning sites that can act simultaneously. The calculation is also complicated by the fact that both the FLL rigidity and the elementary force $f_p$ are functions of temperature and magnetic field. It is necessary to understand how the FLL rigidity limits the number of pinning centers that can contribute their full pinning force. In other words, the main complication is a result of the existence of a flux-line lattice (due to fluxoid repulsion) and not just flux lines. The microstructure-critical current relationships have been recently reviewed by Kramer (5) and we will mention only two of the most prevalent ideas on the summation problem.

Labusch (6) has developed the so-called point-pinning approach. He considers a dilute system of point-pinning centers randomly distributed in the FLL. $F_p$ is found to be proportional to $n f_p u_p$, where $n$ is the pin density, and $u_p$ is the maximum displacement of the surrounding FLL relative to a single point pin. Since $u_p = f_p / C_{eff}$, where $C_{eff}$ is a term containing the elastic constants of the FLL, the volume-pinning-force density is

$$F_p = \frac{N_A}{C_{eff}} f_p^2$$

(6)

where $N_A$ is the area density of flux-pinning centers. The same result was found by considering the power loss of the FLL moving through point pins (2). To date, this model has not been able to predict the microstructural dependence of $F_p(h)$ observed experimentally for dense systems of pinning centers usually found in commercial superconductors, but it has been used successfully to determine $F_p$ for dilute pinning systems (3,7).

A method proposed by Pippard (8) and used by Kramer (9) (denoted the line-pinning model) utilizes the shear capability of the FLL at high fields. The resistance to shear of the FLL ($C_{66}$) decreases as $(1 - b)^{1/2}$ for $b < 1$ ($b = B/B_{C2}$) and
$f_p$ decreases as $(1 - b)$. Therefore, at some point as $b$ increases FLL shear will occur. This will allow an increasing number of pinning centers to interact with flux lines and will result in an increase in $F_p$ as $b$ increases. The model, which requires a certain density and nonrandomness of pinning centers, predicts a strong dependence of $F_p$ on the microstructure in the low-field regime and a microstructure-insensitive $F_p$ in the high-field regime. This model has also been applied successfully to experiments (3,5,9).

Most of the radiation-damage experiments have not been performed in a manner that allows comparison with either of the above models. Proper application requires knowledge of $F_p$ as a function of reduced magnetic flux density $b = B/B_{c2}$ and $T$. In most of the experiments performed on high-field superconductors (and therefore technologically interesting), $B_{c2}$ was too large to measure and changes in $b$ during irradiation could not be determined. This limits the amount of fundamental information available from these experiments. Furthermore, to determine $F_p$ for a particular defect from the experimental $F_p$, it is necessary to have only one type of defect present or to change only one defect density during the experiment.

Many experiments show a "peak" or increase in $F_p$ as $b$ approaches unity. The interpretation of this peak is the subject of controversy (3), but it appears that, as in the summation process, the peak is related to the rigidity of the FLL. As the FLL becomes less rigid, more pinning centers can be contacted by the flux lines and the pinning force increases. It is beyond the scope of the present review to discuss the many examples of peak effect that appear in the experimental data which will be presented.

A concise, coherent understanding of the effects of radiation is difficult to present because of the array of variables in the experiments, i.e. kind of superconducting material (elemental, alloy, and compound), irradiating particle (neutron, electron, and ion), irradiation temperature (generally < 30 K and room temperature), and initial metallurgical condition (such as single crystal, cold-worked, second-phase precipitates, and disordered). The current understanding of the radiation effects will be presented for the three kinds of superconductors, and the roles of irradiation temperature and metallurgical condition will be presented for each. The discussion will be primarily on the results of neutron irradiations, since the majority of the
Experiments involve the use of neutron irradiation because of the technological implications and because the neutrons produce larger flux-pinning changes than ion or electron irradiation. Previous review articles (10-12) point out that, although the electronic properties of the superconductor are affected by irradiation which results in a change in $T_c$, especially in the compound superconductors and $H_c2$, the main feature of the radiation in elemental and alloy superconductors is to alter the magnetic flux pinning due to microstructure changes introduced in the specimen. It is felt that, for each type of superconductor, radiation effects are fairly well understood phenomenologically, i.e. good qualitative agreement with theory and a degree of predictability, but considerable work remains to be done to obtain an understanding of the various elementary fluxoid-defect interactions.

**ELEMENTAL SUPERCONDUCTORS**

The best characterized radiation-damage experiments, in terms of single crystals, impurity levels, single type of defect present, and defect-fluxoid calculations, have been performed in the elemental superconductors. As a result, a semiquantitative understanding of the effects of defects on the elemental superconducting properties has developed.

A decrease in $T_c$ in Nb (up to 0.14 K from an initial 9.25 K) as a function of the change in electrical resistivity $\rho$ has been measured after low-temperature n (13) and d (14) irradiations. A theoretical treatment (15) based on the BCS theory yields an expression that predicts a decrease in $T_c$ due to the scattering from the radiation-induced defects which reduces the anisotropy of the interaction energy between electrons. The theoretical expression

$$\Delta T_c \propto \rho \ln \rho$$

(7)

fits the experimental data (10). $T_c$ for the elemental superconductors is only slightly affected by radiation, since the maximum value of $\Delta T_c = -0.14$ K is small for a large resistivity change of 2 $\mu$-cm, which corresponds to approximately one-half the maximum defect concentration. Annealing the sample that was n-irradiated at 4.6 K (which results in defect annihilation and clustering) led to a nonlinear change in $T_c$ with $\rho$, which indicates that $T_c$ is not only a function of $\rho$ but also depends on the detailed scattering behavior of the defects present.
The linearity and the magnitude of the increase in $H_{c2}$ due to the increase of radiation-induced resistivity are in good agreement with the theoretical predictions of Eqs. (3) and (4). Illmaier et al. (16) irradiated Nb at 4.5 K with 3-MeV electrons. $H_{c2}$ increased proportional to $\tau$ during irradiation, but annealing led to a nonlinear change in $H_{c2}$ with $\tau$. Significant changes in $H_{c2}/\tau$ versus $T$ were seen at the annealing stages, i.e. where the defect structure is altered, which indicates that $H_{c2}$ and $\tau$ depend differently on the scattering behavior of various defect configurations. Using Eq. (4), the theoretical value of $H_{c2}/\tau$ after 5 K fast-neutron irradiation was calculated to be only 4% higher than the experimental value of $H_{c2}$ at 4.5 K. The values of $H_{c2}/\tau$ after 4.5 K e- (16) and 70°C n-irradiation (18) (1.68 and 1.53 Oe/n -cm, respectively) are quite close to the present value. The similar values (in view of the different types of defects present after the three irradiations) indicate that the change in $H_{c2}$ does not depend on the detailed scattering behavior as discussed above.

Tsubakihihara (19) irradiated V with neutrons at 70°C and observed no change in $H_{c2}$. This is tentatively explained as the result of a balance between the increase in $H_{c2}$ due to an increase in $\tau$ and the decrease in $H_{c2}$ due to the observed decrease in $T_c$ (3°). High-purity Nb irradiated at 70°C to $3 \times 10^{11}$ n/cm had an increase in $H_{c2}$ of 100 Oe (4°) (20). Similarly, a 20 Oe increase (3°) was observed in V after the same irradiation (21). The much higher initial $H_{c2}$ in Tsubakihihara's V at 4.2 K (1350 Oe (19), 750 Oe (21)) is probably due to interstitial impurities that can eliminate $H_{c2}$ increases due to irradiation. This lack of change in $H_{c2}$ with dose has been observed in Nb with interstitial impurities irradiated with fast neutrons at 5 K (22). Under identical irradiation conditions, it has been shown that in high-purity Nb the increase in $H_{c2}$ from $\tau$ dominates the decrease in $H_{c2}$ from the small $T_c$ decreases.

Well-controlled experiments have been performed on the flux pinning of defects introduced in elemental superconductors under different irradiation conditions. The capability of defects to pin fluxoids is a function of their size and spatial distribution, since, for significant pinning, the defect size must be of the order of $\xi$ (~340 Å in Nb at 4.2 K). This is clearly seen in Fig. 3, which shows the pinning-force density after 4.5 K irradiations of Nb with electrons (21) and fast neutrons (24) to equal defect concentrations, as measured by the change in $\tau$. As mentioned
in the Introduction, comparisons of the experiments should be made at an $H$ below the peak in $F_p$. Electron irradiation produces isolated defects in the form of Frenkel pairs uniformly distributed over distances less than $\pi$ and results in little flux pinning. Fast-neutron irradiation produces defect cascades ($\sim$50-200 Å in diameter) that are of sufficient size to pin the fluxoids. The unirradiated $H_{c2}$ was the same for the two experiments (2500 Oe at 4.5 K), and $\Delta H_{c2}$ was the same for different types of irradiation to within the limits of Eq. (4). The small amount of pinning that was observed after electron irradiation was not due to a direct interaction between the fluxoids and individual Frenkel pairs, since, for a homogeneous defect distribution, every interaction would be cancelled by an equal interaction of opposite sign and no net pinning force would result. The pinning was explained as being due to fluctuations in the defect density, resulting in a less than perfectly homogeneous distribution. However, the surface could be important in explaining this low $F_p$.

Measurements have been made recently by Brown (17) and Brown et al. (25) on the pinning forces in Nb after fast-neutron irradiation at 4.6 K. Figure 4a shows the change in volume-pinning-force density $F_p$ as a function of dose and field. Changing the superconducting properties locally will alter the Ginzburg-Landau free energy that describes the properties of a type-II superconductor (2). Calculations were made that assumed the elementary interaction force between a defect cascade and a fluxoid is predominantly caused by the magnetic pinning interaction (2). This is caused by the difference in the magnetic energy of flux lines in the defect cascades and the unirradiated crystal, which have different superconducting parameters ($\kappa$ and $H_c$). Considering only the term due to a change in $\kappa$ (and not the term due to a change in $H_c$) leads to a change in the interaction energy $\Delta E_\kappa$:

$$\Delta E_\kappa = V_{\text{ca}} \mu_0 H^2 \frac{\kappa}{\gamma} f(b)$$

(8)

where $V_{\text{ca}}$ is the cascade volume, $f(b)$ is a function of $b$ ($=\pi/\gamma H_{c2}$) of order unity at low $b$ and $\mu_0$ is the permeability constant. The values of $V_{\text{ca}}$ for different neutron energies are, unfortunately, not well known. The use of reasonable estimates of parameters such as saturation resistivity and defect concentration within a cascade led to a $\Delta E_\kappa$ of between 0.005 and 0.17 eV for cascade diameters from 61 to 206 Å that resulted from neutron energies between 0.2 to 7 MeV. These values were used with the experimental neutron
energy spectrum to calculate \( N_A f_p \), where \( N_A \) is the area density of defects, and \( f_p \) is the maximum interaction force. In Fig. 4b, these values are shown to agree with the low-dose experimental values that were determined using the previously discussed statistical model of Labusch (6) [Eq. (6)]. No more than an order of magnitude agreement can be inferred from these results, since uncertainties remain in the calculation of the cascade volume and the reversible magnetization curves required to calculate the FL elastic constants. The calculations were performed at low doses, which resulted in small defect concentrations. The low-temperature neutron irradiations (17,24) show a rapid increase in \( f_p \), a saturation, and a decrease as the dose increases. This is explained as being due to overlap of the damaged regions, which weakens the individual cascade-fluxoid interaction.

Fig. 3. Comparison of Volume-Pinning-Force Density \( f_p \) in Niobium Irradiated with Electrons and Neutrons at Low Temperature to Equal Defect Concentrations.

Sekula (20) and Kernohan and Sekula (26) investigated single crystals of Nb irradiated at ambient temperature (60°C) as a function of dose up to \( 6 \times 10^{19} \) n/cm². Electron microscopy has shown that the size of the resultant radiation-induced dislocation loops varies between 30 and 1400 Å and depends strongly on interstitial impurity content (27). It is expected that loops of this size would lead to considerable flux pinning but would result in a small increase in \( \Delta P \) and therefore \( H_{c2} \). This was observed, and no evidence for a saturation or decrease in flux pinning at higher doses has been observed up to these high dose levels. This is reasonable since at this temperature the defects are mobile (28), and the dislocation loops will grow in size and result in stronger pinning. Preliminary results on fast-neutron irradiation in V at 40°C show a behavior similar to Nb (21).

Experiments have been performed in which the oxygen impurity level in Nb was varied to change the size distribution of the loops that result after 70°C neutron irradiation (27).
The nucleation of loops at oxygen sites is inferred from the increase, by a factor of 20, in the number of "black spots" and loops observed in the TEM for equivalently irradiated samples containing 10 and 900 wt ppm oxygen. Also, a decrease in loop size occurs so that the loop area is approximately constant. The results in Fig. 5 show that the increase in the elementary pinning force \( f_p \) due to the larger loops in the 10-ppm sample dominates the decrease in \( f_p \) due to the decreased loop density. The elementary pinning force \( f_p \) of a dislocation loop has been calculated (29), and the volume pinning force \( F_p \) can be better fit by the quadratic summation of the \( f_p \)s of Labusch [Eq. (6)] than by a linear summation proposed by Dew-Hughes (30), which neglects the rigidity of the FLL.

Thermal-neutron irradiation of V at 5 K by Sekula led to a small increase in \( F_p \) (\( \sim 15\% \)) after \( 2 \times 10^{18} \) n/cm\(^2\) (31). The damage that resulted from thermal-neutron irradiation is due to low-energy events which result in a uniform distribution of defects similar to electron irradiation. The 50% recovery of the \( F_p \) increase after a 60 K anneal is similar to resistivity recovery after a similar irradiation. Preliminary
investigations of technetium irradiated with fast neutrons at 5 K show a behavior similar to that of Nb as \( F_p \) increased and saturated at \( 3 \times 10^{17} \) n/cm\(^2\) (22).

Tsubakihara et al. have performed a thorough investigation of the flux pinning by dislocations in single crystal V (32) and V irradiated in a reactor at 70°C up to 8 \( \times 10^{18} \) n/cm\(^2\) (E > 0.1 MeV) (19). A model is proposed for flux pinning based on an interaction between a random distribution of pinning centers and "flexible" flux lines. The model results in a good experimental fit for the unirradiated sample with a normalized pinning-force density function

\[
\frac{J_{cH}}{(J_{cH})_{max}} = 4h^{1/2}(1-h^{1/2})
\]

which gives a peak in \( J_{cH} \) at \( h = H/H_{c2} = 0.25 \) and is different from the frequently observed \( h^n(1-h)^m \) dependence. Neutron irradiation results in about a factor of two increase in \( J_{cH} \). The normalized \( J_{cH} \) for V irradiated and measured at different temperatures and for V irradiated to different doses and measured at the same temperature fall on the same reduced field curve as described by Eq. (9). Therefore, if the change in \( J_{cH} \) due to the small change in \( T_C \) and \( H_{c2} \) can be ignored, the conclusion is that the n-irradiation at 70°C did not change the nature of the flux-pinning mechanism but simply increased the pinning-site density. As mentioned previously, the unirradiated V probably had a high oxygen content, and applying the results of similarly irradiated high oxygen Nb (27), it is reasonable to suggest that the irradiation increased the loop density, which was observed.

Van der Klein et al. (18) irradiated Nb with an estimated 200-wt ppm interstitial-impurity content at reactor ambient temperature up to 1.5 \( \times 10^{20} \) n/cm\(^2\) and analyzed the results by considering flux pinning at the surface. A surface oxidation process (anneal a few minutes in \( O_2 \) at 400°C) greatly
reduced the magnetic hysteresis of the unirradiated sample, which was attributed to elimination of flux pinning by the surface. The change in $H_{c2}$ with dose was similar to that found by Sekula (20), but the increase in magnetic remanence was at least a factor of five less. The samples were oxidized after irradiation, which resulted in a decrease in $H_{c2}$ to the unirradiated value, as expected after a 400°C anneal (17,28). However, they claim that the flux pinning centers were not affected by the oxidation process by comparing the hysteresis after annealing in oxygen and in vacuum and by electron microscope observation. This argument is not convincing as previous experimental work indicates that defect motion is expected at this temperature (28) which would change the dislocation size distribution and explain the large differences in magnetic remanence changes with dose. Also, the considerably larger interstitial impurity content (200 wt ppm versus 10 wt ppm) would result in a larger number of small loops and, therefore, result in less flux pinning (27).

Freyhardt et al. (33,34) have investigated the effect of voids on flux pinning in Nb and Nb alloys. This system is especially appealing since the size and number distribution of the voids can be well characterized by electron microscopy of simultaneously irradiated samples. The results of high-purity Nb foils irradiated with 3.5-MeV Ni$^+$ ions at 800°C are shown in Fig. 6. The increase in critical current by more than a factor of 100 was due to the voids distributed in a surface layer $\approx 10^6 \AA$ thick. The void size (30-500 Å) and density (up to $2 \times 10^{15}$ cm$^{-3}$) depended on the irradiation conditions and the metallurgical variables, e.g. gaseous impurity content. The voids are usually randomly distributed. However, in materials with a preirradiation high oxygen content, void ordering is observed and a regular void lattice is developed (35). A peak in $J_c$ vs $H$ is observed in this case and appears to be due to a matching of the void and fluxoid spacing. If the fluxoid spacing is some multiple of the void spacing, it is possible for all the fluxoids to be at a void simultaneously without compression of the FLL. This means more fluxoids are being pinned by voids and $F_p$ increases. Fluxoids that are parallel to the surface and thus to the irradiated layer are pinned more effectively than in the perpendicular case. The experimental results were compared with theoretical predictions using the statistical theory (6).

The main contribution to the elementary interaction between the void and fluxoid was from the core interaction (2)

$$\Delta E_c = \frac{\mu H^2}{2} - \frac{\sigma_c}{2} V_{\text{void}}$$

(10)
The statistical theory predicts $J_C$ for the field parallel to the surface that agrees with the experimental value. A comprehensive study of the temperature and field dependence of the volume pinning force is in progress.

Using the above results, a comparison can be made of the pinning strength of different defect configurations of the same size in Nb. A 150 Å cascade (the result of a 4.6 K neutron irradiation) has an elementary interaction force $f_p \approx 4 \times 10^{-13}$ newton (N) that is thought to be primarily due to the magnetic interaction (17). A 150 Å dislocation loop (observed after a 70°C neutron irradiation) has $f_p \approx 23 \times 10^{-13}$ N or six times the cascade $f_p$ and is due to strain-field interactions (27). However, due to the defect mobility and annihilation at 70°C, at low doses the flux pinning per incident neutron in high-purity Nb is about eight times larger for a 6 K irradiation than for a 70°C irradiation. A 150 Å void (observed after a 3.5-MeV Ni+ irradiation at 900°C) has $f_p \approx 7 \times 10^{-13}$ N, considering the core interaction (33).

ALLOY SUPERCONDUCTORS

Most of the radiation-damage experiments in the elemental superconductors had one type of defect as the predominant flux pinning mechanism and could, therefore, yield information on the fundamental fluxoid-defect interaction. The irradiation experiments on alloy superconductors are frequently in material of engineering interest (high unirradiated $J_C$) and usually have more than one species of defect present, such as dislocations, cell walls, precipitates, and defect cascades,
or clusters. Separating unambiguously the contribution of each is difficult and had not, to date, been performed successfully. The problem in the radiation-damage experiments is to determine if the radiation-induced defects are adding to the flux pinning (i.e. summing with the existing defects) or affecting the properties of the material in a manner that results in weaker flux pinning.

A considerable number of irradiations of alloy superconductors, primarily NbTi, have been performed as a result of technological interest. All large superconducting magnets (for accelerators and bubble chambers) use NbTi because of the high $H_{c2}$ ($\sim 12 \, T = 120 \, kG$ at 4.2 K). The salient features of the experiments are that $T_c$ is changed very little by irradiation, and the irradiation-induced changes in $J_c$ and $F_p$ are quite sensitive to the metallurgical structure in the unirradiated material. Using the unirradiated $J_c$ ($J_{co}$) as a metallurgical yardstick, it is generally observed that $J_c$ will increase with irradiation for low $J_{co}$ material and decrease for high $J_{co}$ material, with the magnitude of the decrease larger for larger $J_{co}$.

The radiation-damage experiments in NbTi are summarized in Tables 1-3 in which $\Delta J_c$ is the change in $J_c$ due to irradiation ($\Delta J_c = J_c - J_{co}$). Most of the neutron-irradiation experiments are consistent, and the main features of the experiments can be understood by an inspection of the work by Soell et al. (36) who irradiated Nb-66 at.% Ti with reactor neutrons at 4.5 K. The $J_{co}$ for samples prepared by cold working wire down to 11 μm diameter and annealing for various times and temperatures ($T > 380^\circ C$) are shown in Fig. 7a. It has been shown that the high dislocation density within the cell walls is the predominant pinning mechanism in cold-worked NbTi (48,49). $J_c$ is increased by cold-working and by low-temperature ($T < 400^\circ C$) anneals that produce "fully effective" pinning centers, i.e. give the smallest dislocation-free cell structure. Annealing above 400°C results in cell growth, and, as the cells grow, the total wall area decreases and thus $J_c$ decreases. Electron microscopy has shown that $J_c$ is proportional to $1/d$ (400 Å < $d$ < 1000 Å), where $d$ is the cell size (48).

Soell has shown that the value of the reduced field at which the maximum pinning occurs, $b$ ($F_{p_{max}}$), depends on the dislocation wall density (50). Samples with a low wall density and therefore low $J_{co}$ have low values of $b$ ($F_{p_{max}}$). It was seen that $b$ ($F_{p_{max}}$) and $J_{co}$ were parameters that
Table 1. Low-Temperature Neutron-Irradiation Effects in NbTi

<table>
<thead>
<tr>
<th>Reference</th>
<th>Dose (n/cm², E = 0.1 MeV) and Temperature</th>
<th>Sample</th>
<th>( J_{CO}(\Lambda/cm²) ) at 4.2 K and 4 T</th>
<th>Results (% ( \Delta J/J_{CO} ))</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>36</td>
<td>3.6 x 10¹⁶</td>
<td>Nb-66 at.7 Ti</td>
<td>7 x 10¹⁹</td>
<td>+20</td>
<td>See text.</td>
</tr>
<tr>
<td></td>
<td>4.6 K</td>
<td></td>
<td>1.7 x 10¹⁰</td>
<td>0</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>1.5 x 10⁸</td>
<td>-10</td>
<td></td>
</tr>
<tr>
<td>37</td>
<td>up to 7.5 x 10¹⁵</td>
<td>Nb-66 at.7 Ti</td>
<td>1.1 x 10⁵</td>
<td>-50</td>
<td>( \Delta J_{C} ) linear with dose</td>
</tr>
<tr>
<td></td>
<td>4.6 K</td>
<td></td>
<td>at ( H = 2.7 ) T and 5.3 K</td>
<td></td>
<td>70% recovery by 270 K</td>
</tr>
<tr>
<td>38</td>
<td>up to 3 x 10¹⁶</td>
<td>Nb-44 at.7 Ti</td>
<td>6 x 10⁸ at 0.8 T</td>
<td>-8</td>
<td>50% recovery by 270 K</td>
</tr>
<tr>
<td></td>
<td>6 K</td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>39</td>
<td>up to 9 x 10¹³</td>
<td>Commercial</td>
<td>1.8 x 10⁵</td>
<td>-11</td>
<td>Different dose dependence for single and MF</td>
</tr>
<tr>
<td></td>
<td>77 K</td>
<td>NbTi</td>
<td>2 x 10⁵</td>
<td>-25</td>
<td></td>
</tr>
<tr>
<td></td>
<td></td>
<td>NbTi (MF)¹</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>40</td>
<td>6 x 10¹⁵</td>
<td>Nb 44 at.7 Ti</td>
<td>-1 x 10⁵</td>
<td>-21</td>
<td>( \Delta T_C = -0.5 ) K</td>
</tr>
<tr>
<td></td>
<td>27 K</td>
<td>(MF)¹</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

¹MF is a multifilament composite.
Table 2. Ambient Reactor Temperature (~60°C) Neutron-Irradiation Effects in NbTi

<table>
<thead>
<tr>
<th>Reference</th>
<th>Dose (n/cm²)</th>
<th>Sample</th>
<th>$J_{CO}$ (A/cm²) at 4.2 K and 4 T</th>
<th>Results ($\Delta J/J_{CO}$)</th>
<th>Comments</th>
</tr>
</thead>
<tbody>
<tr>
<td>41</td>
<td>$4.8 \times 10^{19}$ ((E &gt; 1 \text{ MeV}))</td>
<td>Nb-52 at.% Ti</td>
<td>&lt;10³ at 0.3 T, 5 x 10³ at 0.7 T</td>
<td>+30</td>
<td>Measured by ac losses</td>
</tr>
<tr>
<td>42</td>
<td>up to $6 \times 10^{19}$ ((E &gt; 1 \text{ MeV}))</td>
<td>Nb-64 at.% Ti (MF)</td>
<td>$6 \times 10^4$</td>
<td>-19</td>
<td>$\Delta J_{CO}/J_{CO}$ linear with dose</td>
</tr>
<tr>
<td>43</td>
<td>up to $1.3 \times 10^{18}$ ((E &gt; 0.1 \text{ MeV}))</td>
<td>Nb-48 at.% Ti</td>
<td>1-8 x 10⁴</td>
<td>No systematic change. Decreases for high $J_{CO}$</td>
<td></td>
</tr>
<tr>
<td>44</td>
<td>$3.5 \times 10^{18}$ ((E &gt; 0.1 \text{ MeV}))</td>
<td>Nb-66 at.% Ti</td>
<td>$3 \times 10^4$</td>
<td>+100</td>
<td>Inconsistent with other experiments</td>
</tr>
<tr>
<td>Reference</td>
<td>Sample</td>
<td>Irradiating Particle, J&lt;sub&gt;co&lt;/sub&gt;(A/cm&lt;sup&gt;2&lt;/sup&gt;) at Dose and Temperature</td>
<td>Results (ΔJ/J&lt;sub&gt;co&lt;/sub&gt;), %</td>
<td>Comments</td>
<td></td>
</tr>
<tr>
<td>-----------</td>
<td>---------</td>
<td>-------------------------------------------------</td>
<td>-------------------------------</td>
<td>----------</td>
<td></td>
</tr>
<tr>
<td>45</td>
<td>Nb-58 at.% Ti</td>
<td>50-MeV deuterons 3.5 x 10&lt;sup&gt;16&lt;/sup&gt;/cm&lt;sup&gt;2&lt;/sup&gt; 10 K</td>
<td>8 x 10&lt;sup&gt;9&lt;/sup&gt;</td>
<td>0</td>
<td>See text.</td>
</tr>
<tr>
<td>46</td>
<td>Nb-66 at.% Ti</td>
<td>3.1 MeV protons 1 x 10&lt;sup&gt;17&lt;/sup&gt;/cm&lt;sup&gt;2&lt;/sup&gt; 25 K</td>
<td>1.5 x 10&lt;sup&gt;5&lt;/sup&gt;</td>
<td>-19</td>
<td>See text.</td>
</tr>
<tr>
<td>47</td>
<td>Nb-60 at.% Ti</td>
<td>13-15 MeV protons 7 x 10&lt;sup&gt;17&lt;/sup&gt;/cm&lt;sup&gt;2&lt;/sup&gt; &lt;30 K</td>
<td>1.7 x 10&lt;sup&gt;5&lt;/sup&gt;</td>
<td>-3</td>
<td>Similar change after 10&lt;sup&gt;18&lt;/sup&gt;/cm&lt;sup&gt;2&lt;/sup&gt; at 150 K</td>
</tr>
<tr>
<td>48</td>
<td>Nb-61 at.% Ti</td>
<td>15 MeV deuterons 1 x 10&lt;sup&gt;17&lt;/sup&gt;/cm&lt;sup&gt;2&lt;/sup&gt; 30 K</td>
<td>2.3 x 10&lt;sup&gt;4&lt;/sup&gt; at H = 3.5 T and T = 7 K</td>
<td>-60</td>
<td>2% decrease in T&lt;sub&gt;c&lt;/sub&gt; and 13% decrease in H&lt;sub&gt;c2&lt;/sub&gt;; J&lt;sub&gt;c&lt;/sub&gt; and H&lt;sub&gt;c2&lt;/sub&gt; recovered 90% by 300 K.</td>
</tr>
</tbody>
</table>
determined the effect of irradiation. This is reasonable since the maximum pinning force should be most sensitive to radiation-induced changes in the pinning force. The changes in $J_c$ after irradiation are shown in Fig. 7b as a function of $f(F_p, \text{max})$. The increase in $J_c$ with irradiation for highly annealed sample 4 is due to the introduction of flux-pinning defect cascades. Soell et al. (37) attribute the pinning in the unirradiated specimen to the dislocations in the cell walls and explain the decrease in pinning in the high $J_{co}$ samples to an increase in the effective defect density for pinning within the cell cores. If the relative change in the defect structure in the cell walls is small, the increase in defect density in the relatively defect-free cell cores decreases the difference in the defect structure of the walls and cores. This decreases the difference in effective pinning density of the cell walls and the cell cores. This "shorting out" of the cell walls depends on the initial cell size and results in a decrease in flux pinning.

An experiment has been suggested to test this cell-wall "shorting out" model (51). It is thought that, if this model is correct, the best experiment to perform is one which eliminates all flux pinning except that due to the cell walls. We will irradiate cold-worked, high-purity Nb at 6 K with thermal neutrons. Low-temperature irradiation by
thermal neutrons results in a uniform distribution of immobile point defects. These defects are weak flux pinners (31), and, if the model is correct, $J_c$ should initially decrease with dose. No initial decrease has been observed in the elemental superconductors, although all experiments have been performed with either single crystals that have weak flux pinning or irradiated under conditions which resulted in defects capable of flux pinning.

It is clear that sample 5 in Figs. 7a and 7b does not follow the rule that the maximum $J_{c0}$ leads to maximum $J_c$ decrease with dose. Sample 5 was made from an alloy with a higher Ti content and contained α-phase Ti-rich precipitates. The precipitates as well as the cell walls acted as pinning centers in the unirradiated sample, and the results indicate that the irradiation did not affect the pinning characteristics of the precipitates. The results of liquid-helium temperature irradiations are consistent and permit a fairly accurate prediction of the effect on $J_c$ of fast-neutron irradiation and subsequent anneals in high $J_{c0}$ material. This is the sort of engineering information required for magnet builders for future accelerator or fusion-reactor magnets.

The ambient reactor temperature (∼60°C) irradiations of NbTi have little technological relevance and suffer from more metallurgical questions (such as radiation-induced solute segregation or precipitation) than low-temperature irradiations as a result of defect mobility at the irradiation temperature. For example, Soell, et al. (36) observed that the Ti-rich precipitates were resistant to neutron irradiation at 5 K, whereas, Tsubakihara et al. (43) attributed the decrease in flux pinning after 60°C neutron irradiation to the break up and re-solution of the Ti-rich precipitates. An additional example of defect mobility affecting the flux pinning is an ambient temperature irradiation of Nb-10 at.% Mo, which showed an increase in $F_p$ due to a matching in the FLL spacing to an integral number of periods of the spinodal decomposition product (52). It was suggested that the kinetics of the spinodal decomposition could be greatly enhanced by the mobile radiation-induced defects. Resistivity measurements on 6 K fast-neutron-irradiated NbTi show 75% recovery by 350 K with the recovery stage III at 250 K (stage III is at 200 K in Nb) (28). Therefore, during an ambient temperature irradiation, the defects are mobile and will be in the form of defect clusters or dislocations after the irradiation. The considerably different flux-pinning properties of these structures and defect cascades which result after a helium temperature irradiation, as
discussed in the Elemental Superconductors section, show that the measurements after ambient temperature irradiations of alloys will be of little help in predicting radiation effects in a magnet operated below 10 K.

Wohlleben (46) made a thorough investigation of the influence of 3.1-MeV proton irradiation at 25 K on Nb-66 at.% Ti. $J_c$ was measured as a function of proton dose, magnetic field ($1 \text{T} \leq B \leq 8 \text{T}$), and temperature ($2.5 \text{K} \leq T \leq T_c$). Irradiation resulted in a $T_c$ decrease of 0.17 K ($T_{c0} = 8.85 \text{K}$) and an almost uniform degradation of 19% over the entire pinning-force density surface $F_p(B,T)$. A $J_c$ recovery of 60% was noted after annealing 1 h at 285 K. The results were found to obey a scaling law

$$F_p(B,T) = J_c B \cdot 4F_{p \text{ max}} h(1-h) \quad (11)$$

$$F_{p \text{ max}} = A H_{c2}(T)^2 \quad (12)$$

where $A$ is a constant that depends on the microstructure and composition of the material. Similar scaling laws have been found for many high-field superconductors (9). The fact that the $h$ dependence of $F_p$ does not change during irradiation shows that the mechanism responsible for the pinning has been influenced relatively little by the irradiation-induced defects. It is expected that the defects resulting from 3.1-MeV proton irradiation will be primarily due to low-energy events which are the result of the coulomb interaction. These events will produce isolated Frenkel pairs that are immobile at the 25 K irradiation temperature (28) and have been shown to be weak flux-pinning sites (23,31). If the dislocation structure of the cell walls is the predominant pinning mechanism, as discussed previously, it is expected that the introduction of a uniform distribution of isolated defects in the cell cores will not act as pinning sites but rather decrease the cell-wall pinning. In other words, as observed, the irradiation-induced defects do not change the $h$ dependence of $F_p$ that is characteristic of flux pinning by cell walls but only act to decrease the strength of the cell-wall pinning as represented by $F_{p \text{ max}}$. Inserting the experimental results into Eqs. (1) and (2) shows that 70% of the observed $F_p$ degradation is due to a decrease in $A$ [Eq. (12)] and 30% from a decrease in $H_{c2}(T)$ due to the decrease in $T_c$.

Seiht et al. (45) irradiated Nb-58 at.% Ti with 50-MeV deuterons at 10 K up to a dose of $3.5 \times 10^{16}/\text{cm}^2$ and observed no change in $J_c$. The number of defects $n$ produced by ions of mass $M$, energy $E$, and charge $Z$ in the MeV energy range is (53)
and the defects are in the form of isolated Frenkel pairs that result from low-energy events. A 50-MeV d\(^+\) will produce \(\sim 40\%\) as many defects as a 3.1-MeV p\(^+\). Therefore, 50-MeV d\(^+\) to a dose of \(3.5 \times 10^{17}/\text{cm}^2\) would result in 15% as many defects as Wholleben's 3.1-MeV p\(^+\) irradiation to \(1 \times 10^{17}/\text{cm}^2\). Assuming a linear change in \(J_c\) with the number of defects, the irradiation would give a 37% decrease for this experiment which is less than the measurement error, and, therefore, the lack of change in \(J_c\) is reasonable.

**COMPOUNDS**

Recently, considerable interest in radiation effects in compound superconductors has been generated for technological and fundamental reasons. Since \(H_{c2}(4.2\,\text{K})\) for NbTi is 12 T, all superconducting magnets that require a field greater than this must use a compound superconductor such as Nb\(_3\)Sn with \(H_{c2}(4.2\,\text{K})\) \(\sim 25\,\text{T}\). If these materials are to be used in magnets subjected to a radiation environment it is necessary to know the effects of radiation on the superconducting properties as in the case of NbTi magnets. A recent review discusses the radiation effects in the various magnet components (superconductor, stabilizer, insulator and structural material) and the anticipated neutron spectrum and flux level at the magnet (54). Fundamental interest was initiated by the fact that \(T_c\) is a sensitive function of the degree of long-range crystallographic order (LRO) in the sample (55), and it was observed that neutron irradiation introduced sufficient disorder to lower \(T_c\) significantly.

Most radiation-damage experiments in compound superconductors have been on the A-15 compounds (A\(_3\)B), which have recently been reviewed (56,57). The structure consists of one B atom on the cube corners and two A atoms along the [100] direction on each face. This results in orthogonal chains of A atoms along the three [100] directions, which are believed to play an important role in the superconductivity of the A-15 compounds. Any break in these chains by the introduction of B atoms on A sites will decrease the LRO and result in a degradation of the superconducting properties. It has been shown that, for compounds in which the B atom is not a transition element, the maximum \(T_c\) is obtained for the compound with the maximum LRO parameter \(S\) (55), i.e. only A atoms at
A sites and B atoms at B sites. In V$_3$Au, $T_c$ increased from 0.7 to 3.2 K as $S$ increased from 0.80 to 0.99. The metallurgical problems involved in preparing A-15 compounds are considerable in view of this high sensitivity to LRO and their extreme brittleness. Recent experiments have resulted in the highest $T_c$ for any superconductor, which is 23 K for a thin film of vapor-deposited Nb$_3$Ge (58).

A number of experiments have been performed in which the change in $T_c$ as a function of fast-neutron dose has been measured (59-61). The results of measurements performed after irradiation at 10 K (59) and ambient reactor temperature (60) (Fig. 8) show little dependence of the $T_c$ depression on irradiation temperature. Similar fractional $T_c$ decreases with dose were seen in Nb$_3$Al and Nb$_3$Ga after the 140°C irradiation. The amount of disorder introduced by irradiation depends on the number of replacement events. Since this is a function of the initial damage event and not subsequent defect annealing, the change in $S$ and $T_c$ should be independent of irradiation temperature. The small differences observed can be attributed to the use of different $T_c$ measuring techniques: resistance in Ref. 59 and induction in Ref. 60.

![Fig. 8. Critical Temperature Change $T_c$ in Nb$_3$Sn with Neutron Irradiation.](image)

$T_c$ changes have been measured by Sweedler and Cox (62) in Nb$_3$Al irradiated at 140°C up to $5 \times 10^{19}$ n/cm$^2$ (E > 1.0 MeV) (62). For doses $> 1.4 \times 10^{19}$ n/cm$^2$, no superconductivity was observed down to 1.4 K, the lowest temperature attained. The $T_c$ decreases (similar to Fig. 8) were correlated with decreases in the Bragg-Williams LRO parameter $S$. During the irradiation, the Nb$_3$Al maintained the A-15 lattice structure, but $S$ (as determined by neutron-order-diffraction) and $T_c$ decreased, with $T_c$ a linear function of $S$ for the Nb site. The large amount of localized damage that resulted from a high-energy neutron collision with a lattice atom caused sufficient replacement of Al on Nb sites so as to severely degrade the superconducting properties. The use of expressions of $S$ for the Nb and Al sites and a consideration
of only a simple substitutional order-disorder model led to a determination of the number of Nb and Al site exchanges as a function of dose as shown in Fig. 9. It was seen that a depression in $T_c$ of 2.2 ± 0.2 K occurred for each percent of Nb sites occupied by Al atoms, for 9.6 < $T_c$ < 18.7 K. This depression rate is quite large, and it is speculated that all the A-15's will show this sensitivity to atomic ordering. For a sample of Nb$_3$Al with $T_{co}$ = 18.7 K after 5 x 10$^{19}$ n/cm$^2$, $T_c$ was less than 1.4 K, but a 750°C anneal for 10 min resulted in $T_c$ = 15.9 K and after 80 min $T_c$ = 17.7.

Some preliminary results indicate that the $T_c$ of Laves phase material such as V$_2$Hf is less sensitive to radiation (i.e. less depressed) than the A-15's but the recently discovered PbMo$_x$S$_y$ is more sensitive. After fast-neutron irradiation at 60°C to 2 x 10$^{18}$ n/cm$^2$ (E > 0.1 MeV), the fractional $T_c$ depression was 5% in Nb$_3$Al, 2% in V$_2$Hf, 14% in PbMo$_6$S$_7$, and 12% in SnMo$_5$S$_8$ (63). After reactor neutron irradiation at 140°C to 10$^{19}$ n/cm$^2$ (E > 1 MeV), the fractional $T_c$ depression was 50% in Nb$_3$Al, 10% in V$_2$Hf, and >70% in PbMo$_x$S$_y$ (64).

$J_c$ was measured by Brown et al. (65) in multifilament wires of Nb$_3$Sn with a high $J_{co}$ (1.5 x 10$^6$ A/cm$^2$ at 3.2 T and 4.5 K) that were irradiated at 6 K with fast neutrons to a dose of 1.8 x 10$^{18}$ n/cm$^2$ (E > 0.1 MeV). The fractional changes in $J_c$ (as defined by a 1-nV signal over 7 mm) are shown in Fig. 10. The $J_c$ increases are seen to be greater at larger fields and decreases are observed for H < 0.8 T. As the dose increased, the $J_c$ increase saturated and decreased for higher doses: the saturation dose was largest for the highest field. No $T_c$ changes were measured, and it is estimated that $T_c$ decreased by <1 K at the highest dose (Fig. 8). Approximately
Fig. 10. Fractional Change in $J_c$ after 6 K Fast-Neutron Irradiation.

$F_p = H_c^m f(h,d)$

where the exponent $m$ is $>2$, and $f(h,d)$ is a function of the reduced field $h$ and the defect microstructure represented by $d$ (9). Furthermore, the field dependence can be expressed as

$f(h,d) = A(d) h^n (1-h)^2$

where $n$ is a constant. The dose dependence of the three terms in $F_p$ ($T_c$, $H_{c2}$, and $d$) can be seen by calculating the differential change in $F_p$ with dose (65)

$$\frac{\Delta J_c}{J_{co}} = \frac{\Delta F_p}{F_p} = \left[ C_1 \frac{\partial \ln T_c}{\partial (\phi t)} + C_2 \frac{\partial \ln H_{c2}}{\partial (\phi t)} + C_3 \frac{\partial \ln A(d)}{\partial (\phi t)} \right] \Delta (\phi t)$$

The first term within the brackets is negative, since $T_c$ decreases with dose ($= \phi t$), and corresponds to the deleterious
effect of $T_c$ decreases on the critical current. The third term is positive and corresponds to the increase in cascade pinning with dose [$A(d)$ depends only on microstructure]. However, at high doses saturation occurs, as in the case of Nb, because of overlap of the neutron-damaged regions (17) and this term becomes smaller as a function of dose. Although neither the first nor the third term is a function of applied field, the coefficient of the second term is strongly dependent on $H$; negative at low fields and positive at high fields. The second term corresponds to the change in $J_c$ as a result of the increase in $H_{c2}$ with dose [see Eq. (4)]. If the values of $C_1$, $C_3$, and $C_2(h)$ are of the correct magnitude, $\Delta J_c/J_{c0}$ will be negative at low $H$ and positive at higher $H$, which is the behavior observed in Fig. 10. Moreover, since the magnitude of the positive third term decreases with dose as a result of the saturation effect of overlapping cascades, the field above which a reversal in sign of $\Delta J_c/J_{c0}$ occurs from negative to positive will increase as the dose increases, again as observed. The cascade overlap is seen to be significant after low doses, since a peak is observed at $6 \times 10^{17}$ n/cm$^2$ for $H = 1.23$ T, which is approximately equal to the saturation dose observed in Nb (17). This model could not be quantitatively tested since it was not possible to install a magnet capable of measuring $H_{c2}$ (>14 T). It would be interesting to test this model by measuring a high $J_{c0}$ sample at higher fields or at higher temperatures (and use scaling laws) to determine $H_{c2}$ and $\Delta H_{c2}/\Delta T(t_0)$ and to doses ($\approx 10^{19}$ n/cm$^2$) where $T_c$ decreases would have a significant effect on $F_p$ changes.

The thermal transition current $I_T$, which corresponds to the complete thermal transition to the normal state, was also measured. The transition from the superconducting to the normal state depends on the applied field, current density, and heat generated by the voltage drop across the sample. $I_T$ depends on both thermal and fundamental superconducting properties. The Nb$_3$Sn layers are embedded in a Cu-Sn alloy, and the increase in normal-state resistivity with irradiation and the Wiedemann-Franz law predict that the thermal conductivity of the cladding will decrease with dose. Therefore, $I_T$ will depend on the $F_p$ changes, as discussed above, and on the decrease in thermal conductivity of the cladding as the dose increases. As the capability of the cladding to conduct heat from the Nb$_3$Sn decreases with dose, it is expected that $I_T$ should decrease with dose more rapidly than $J_c$. This is observed as the peak in $I_T$ versus dose occurs at a smaller dose than the peak in $J_c$. The apparent inconsistent increase in $I_T$ during annealing is due to a larger recovery in the thermal conductivity of the cladding ($\approx 60\%$ by 295 K) than the $J_c$ of the superconductor ($\approx 25\%$), resulting in an enhancement of $I_T$. 
The application of these results to fusion magnets is indicated in Fig. 11. This is a compilation by Ullmaier (54) of the results of 6 K neutron irradiations in NbTi (36) and Nb₃Sn (65) and the estimated neutron dose (66) incident on a magnet after 10 years of continuous operation. The neutron spectrum is similar to a fission spectrum (66), and therefore the bulk of the superconductor in the magnet will not show significant detrimental effects of radiation after long periods at the estimated flux.

Measurements have been made by Colucci et al. (67) of Nb₃Sn samples prepared and neutron irradiated at 6 K, with irradiations similar to those given in Ref. 65 except that a warm up to 78 K occurred during the transfer to a 10 T magnet before measurement. Less than 15% recovery in Jₐ was seen at this temperature (65). The increase observed after 5 x 10¹⁷ n/cm² (at 3.2 T) agrees with the data of Ref. 65. Also, ΔJₐ/Jₐ₀ increases as the field increases, and the 50% increase seen at 10 T agrees quite well with an extrapolation of the results in Fig. 10. The ΔJₐ increases after 1.1 x 10¹⁸ n/cm² were less than after 5 x 10¹⁷ n/cm² and do not agree with Fig. 10 (68). Higher dose experiments and measurements that determine the changes in Hₐ with dose and annealing are in progress in an attempt to understand the discrepancies.

It has been observed that grain boundaries are the predominant flux-pinning mechanism in Nb₃Sn with larger Jₐ for smaller grains (69,70). Different fabrication processes (i.e. heat-treatment time and temperature or impurity addition) result in different grain sizes and, therefore, different Jₐ₀. The work in NbTi showed that the change in Jₐ with irradiation was a function of Jₐ₀, which depended on the cell size. In Nb₃Sn, a dependence of the change in Jₐ with irradiation was also seen to be a function of Jₐ₀ which depended on grain size. Nb₃Sn wires made by vapor-phase diffusion have been irradiated at 6 K with reactor spectrum neutrons to 3.9 x 10¹⁸ n/cm².
\( J_{C0} \) was \( \sim 6 \times 10^5 \, \text{A/cm}^2 \) (at 3 T and 4.2 K), which is more than a factor of two lower than the \( J_{C0} \) of Ref. 65. A \( J_c \) enhancement by a factor of 2.5 (1.5 T < H < 5 T) and a \( T_c \) reduction of 0.8 \pm 0.1 K were observed. Approximately half of this enhancement remained after annealing to 250 K. These results are not inconsistent with Ref. 65, since the \( J_c \) after the present irradiation was approximately the \( J_{C0} \) given in Ref. 65. It is expected that \( J_c \) would decrease at higher doses.

No changes in \( J_c \) on Nb\(_3\)Sn wires similar to those used in Ref. 65 were observed by Parkin and Sweedler (72) after reactor neutron irradiations to doses <2 \times 10^{18} \, \text{n/cm}^2 \, (E > 1 \, \text{MeV}) at 60°C, which is in a dose regime where substantial increases in \( J_c \) were observed (Fig. 10) after 5 K irradiation. The large decreases in \( J_c \) after higher doses are attributed to decreases in \( T_c \). Therefore, although the radiation-induced disorder and resultant \( T_c \) decrease are comparable after 6 K and 100°C irradiation (Fig. 8), the different types of radiation-induced structures have substantially different effects on the flux pinning. The results in Nb show that at low doses the defect cascades present after 6 K irradiations are much more effective at flux pinning than the clusters or loops present after 60°C irradiation to an equal dose. It is seen that the same is true in Nb\(_3\)Sn.

Preliminary results indicate that samples irradiated as in Ref. 72 show an increase in \( J_c \) at higher fields (H > 4 T) upon irradiation and decreases in \( J_c \) at large doses (73). Similar samples of Nb\(_3\)Sn showed no change in \( J_c \) (at 4 T) after 14-MeV neutron irradiation at room temperature to a dose of 1 \times 10^{18} \, \text{n/cm}^2 \, (74). Nb\(_3\)Sn wires made by vapor-phase diffusion were reactor neutron irradiated at 80°C up to 5 \times 10^{18} \, \text{n/cm}^2 \, (E > 0.8 \, \text{MeV}) and measured up to 10 T (75). Increases in \( J_c \) by a factor of three were seen after 1 \times 10^{18} \, \text{n/cm}^2 \, with a flattening of the \( J_c(H) \) curve after 5 \times 10^{18} \, \text{n/cm}^2 \, i.e. decrease in \( J_c \) at low H and increase at high H. The low \( J_{C0} \) (\( \sim 4 \times 10^5 \) at 3.5 T and 4.2 K) makes these results consistent with other high-temperature irradiations (61,72). Simultaneous irradiation of Nb\(_3\)Al, Nb\(_3\)(Al,Ge), and V\(_3\)Si showed similar results. The earlier irradiations of A-15 compounds were performed on low \( J_{C0} \) material (76-78). As in NbTi, a dependence of the change in \( J_c \) with irradiation on \( J_{C0} \) was seen. For example, neutron irradiation at 50°C on vapor-deposited Nb\(_3\)Sn, with \( J_{C0} = 0.95 \times 10^5 \, \text{A/cm}^2 \) (at 1.4 T and 4.2 K), showed an increase to 1.3 \times 10^6 \, \text{A/cm}^2 \, after 1.4 \times 10^{18} \, \text{n/cm}^2 \, (E > \?) (76). The same irradiation on a Nb\(_3\)Sn sample with \( J_{C0} = 9.5 \times 10^5 \, \text{A/cm}^2 \, led to a decrease to 0.9 \times 10^5 \, \text{A/cm}^2 \).
Bauer et al. (75) have irradiated $^{235}$U-doped samples of A-15 compounds. Irradiation in a thermal-neutron flux at $80^\circ$C causes fissioning of the $^{235}$U and a high damage rate results from the fission fragments. This is a mixture of isolated defect and fast-neutron type of damage but has been shown at high doses to approximate fast-neutron irradiation (79).

Initial increases in $J_C$ with dose can again be attributed to the relatively low $J_{CO}$, $8 \times 10^9$ A/cm$^2$ (at 3 T and 4.2 K). At intermediate doses, a dramatic flattening of the $J_C(H)$ curve occurs, as was observed in Ref. 65. Irradiation to higher doses resulted in substantial decreases in $J_C$, which is attributed to the decrease in $T_C$ that is significant at these doses. Samples doped with $^{10}$B show similar but considerably less pronounced results because of the damage that results from the (n,$\alpha$) reaction. Both doped samples of Nb$_3$Sn showed decreases in $T_C$ proportional to an increase in the lattice constant. This is consistent with the data of Ref. 62, which attributes the decrease in $T_C$ to an exchange of Nb and Sn atoms that results in a decrease in the IRO and an expansion of the lattice. Damage that resulted from the (n,$\alpha$) reaction with $^{10}$B or the $^{235}$U fission did not increase $J_C$ to a value greater than that after fast neutron irradiation.

The problem with comparing these results with those of Refs. 65 and 72 is that in the latter experiments $J_{CO}$ was larger than the maximum $J_C$ obtained in the present experiment. The low $J_{CO}$ material has a high degree of order, as seen by a high $T_{CO}$, but a nonmaximized flux pinning microstructure and the irradiation simultaneously decreases the first and improves the second. After a dose at which $J_C$ has become large, the deleterious effects of $T_C$ decreases are becoming significant. Thus, it is difficult to predict the irradiation effects in high $J_{CO}$ material from the results on low $J_{CO}$ material.

The change in $T_C$ was measured after 25-MeV oxygen-ion irradiation at 30 K on vapor-deposited Nb$_3$Sn ribbons (80). $T_C$ decreased with dose, and after $1.5 \times 10^{16}$ ions/cm$^2$, $T_C$ had decreased to 4 K from an initial value of 18.5 K. Only 37 of the decrease recovered after a 300 K anneal. The $T_C$ changes are again explained as the result of radiation-induced disordering. The changes in $J_C$ in similar samples were measured after 24-MeV oxygen-ion irradiation at 110$^\circ$C (81). $J_C$ for initially low $J_{CO}$ material ($5 \times 10^5$ A/cm$^2$ at 3 T and 4.2 K) increased approximately 50% after $3 \times 10^{14}$/cm$^2$ and decreased to below $J_{CO}$ after $1.6 \times 10^{15}$/cm$^2$. As discussed in the NbTi results, this type of radiation is expected to result primarily in low-energy events which will produce isolated point defects that will cluster at this irradiation temperature. The average energy transferred in the primary
impact is 300 eV for 24-MeV oxygen ions and 10 keV for a 0.5-
MeV neutron. The small pinning effects for this type of damage
have been discussed. It is felt that the increase in $I_c$ ob-
served at low doses was attributable to the low $J_{CO}$. The
previous results indicate that the decrease in $I_c$ at high doses
is due to the $T_c$ decreases.

Wohlenberg (62) measured the change in $I_c$ of Nb$_3$Sn tapes
irradiated at 100°C with 1-, 2-, and 3-MeV protons and 3-MeV
deuterons. The samples were prepared by diffusion at 975°C
for 4 h and had low $I_{CO} (4 \times 10^{11} \text{A/cm}^2$ at 1 T and 4.2 K).
For this type of irradiation, the defects are produced as a
result of the coulomb interaction. Rutherford scattering
theory gives the ion-scattering cross section $\sigma$ proportional
to $M^2/E$ where $M$, $Z$, and $E$ are the mass, charge, and energy
of the irradiating ion. It was possible to plot the $J_c$
changes for the different irradiating ions as a function of
normalized dose $S_{\text{norm}}$

$$S_{\text{norm}} = \frac{M^2}{E} S$$

The results show that a peak in the $J_c$ ($S_{\text{norm}}$) curve occurs
at the same $S_{\text{norm}}$ for the different irradiations. The $J_c$
changes were then calculated taking into consideration a def-
cet cluster distribution. As discussed previously (65) it
is necessary to consider changes in $T_c$ and $H_{c2}$ as well as the
defect microstructure when analyzing $J_c$ changes. The use of
Eq. (17) for the $T_c$ changes in Nb$_3$Sn after 24-MeV oxygen-ion
irradiation (60) would predict $T_c = 3 K$ (after $1 \times 10^{17}$
$p/$cm$^2$, $E = 1$ MeV), which would have been considered in
the calculations. $T_c$ and $H_{c2}$ changes were not measured nor con-
idered when calculating $J_c$ changes. Therefore, it is felt
that the calculations of the $J_c$ changes based only on flux
pinning by defect clusters is incomplete.

Recent work by Seibt et al (83, 84) on V$_3$Ga and Nb$_3$Sn
irradiated with 50-MeV deuterons up to $4 \times 10^{17}$ d/cm$^2$ below
18 K show behavior similar to that presented in Fig. 10. In-
creases in $J_c$ of up to 75% (at 7 T) were seen in Nb$_3$Sn, with
the greatest increase for the largest field. The increase
saturated below $5 \times 10^{16}$ d/cm$^2$ and $J_c$ decreased for higher
doses. $J_c$ for V$_3$Ga showed a similar change with dose, and the
percentage increase and saturation dose depended on $J_{CO}$.
The change in $T_c$ was much smaller than the previous 25-MeV
oxygen irradiation (80) [as predicted by Eq. (17)] with a 7%
reduction after $1.9 \times 10^{17}$ d/cm$^2$ ($T_{CO} = 17.8 K$).

B-1 compounds [NbN and Nb((1,N)) doped with $^{10}$B and $^{23}$U
have been irradiated at 70°C (85). The changes with irradiation
were considerably less dramatic than for the A-15's with small increases in $H_{c2}$ and the lattice parameter and no changes in $T_c$. $J_c$ decreased in NbN and increased in Nb(C,N), with the largest changes seen at the peak in $J_c(H)$ near $H_{c2}$.

Considerable work has been done in alloys to maximize $J_c$ by cold-work, addition of impurities to form precipitates, and various heat treatments. Methods to increase $J_c$ cannot be applied as easily to the A-15 compounds because of their sensitivity to LRO and severe brittleness. A tension strain only 0.2% will result in cracking (86). The addition of impurities has raised $J_c$, but it is not known if this is due to pinning by impurity precipitates or if the grain growth is inhibited by impurities. Lowering the reaction temperature decreases the grain size and increases $J_c$, but a slow growth rate results. A reaction temperature of 600°C produces small grains of 1-450 Å diameter, but 350 h where required to yield a 2-μm-thick Nb₃Sn layer (69). Reaction temperatures that would yield maximum flux pinning (small grains) would proceed in a prohibitively slow manner. The various methods to enhance $J_c$ are covered in the review (36), and we have mentioned briefly some of the metallurgical problems to permit a better understanding of the radiation effects. The high $J_c$ alloys have been prepared in a manner that will give maximum $J_c$, and it is not surprising to observe that a change in metallurgy in high $J_c$ material by any means (e.g. any type of irradiation) leads to a decrease. However, it is not possible to "maximize" $J_c$ for the compounds because of metallurgical considerations. Therefore, it is reasonable that $J_c$ can be raised even for high $J_{c0}$ material by a process that will introduce flux-pinning sites and not affect LRO or strain. It was seen that low doses of fast neutrons at 6 K produce the required damage (65). Higher doses lower the LRO, and irradiations at higher temperatures and low doses result in weaker flux-pinning sites.

CONCLUSIONS

The effect of radiation on the superconducting transition temperature ($T_c$), upper critical field ($H_{c2}$), and volume-pinning-force density ($F_p$) were discussed for the three kinds of superconducting material (elements, alloys, and compounds).

Elements

Most information on fundamental fluxoid-defect interactions ($F_p$) has been obtained from experiments in elements, since the starting material has a well-characterized nature. Calculated values of $F_p$ for defect cascades, dislocation loops, and voids
agree with experiment. Small $T_c$ and large $H_{c2}$ changes are in good agreement with theory.

**Alloys**

Interpretation of flux-pinning experiments in alloys is difficult due to the presence of more than one type of defect that has the capability of flux pinning (such as dislocations or precipitates and radiation-induced defects). The sign and magnitude of the change in $F_p$ after irradiation is a function of the initial metallurgical condition, as characterized by the unirradiated $F_p$ ($F_{po}$). $F_p$ decreases with irradiation in high $F_{po}$ material and increases in low $F_{po}$ material.

**Compounds**

Although $T_c$ decreases very little with irradiation in the elements and alloys, the $T_c$ of compounds can be substantially reduced as a result of radiation-induced disordering. Changes in $T_c$, $H_{c2}$, and the defect microstructure must be considered when analyzing the $F_p$ changes due to irradiation. As in the case of alloys, the magnitude of the change in $F_p$ with irradiation is a function of the initial metallurgical state. However, even in high $F_{po}$ material, $F_p$ can increase at low doses where $T_c$ decreases are small.

A good phenomenological understanding of the effect of irradiation on the properties of superconductors exists. It does not appear at present that radiation effects in superconductors (for fusion magnets) will be a technological limitation. Work in the future should concentrate on experiments that can determine the fundamental defect-fluxoid interaction for the defect structures resulting from different types of irradiation.

**ACKNOWLEDGEMENTS**


**REFERENCES**

(2) A. M. Campbell and J. E. Evetts, Adv. in Physics, Vol. 21, 1972, p.199.
(17) B. S. Brown, IDMFPS, p.200.
(29) E. J. Kramer, submitted to Phil. Mag.
(31) S. T. Sekula, IDMFPSS, p. 206.
(40) M. Couach, private communication, 1975.
(51) E. J. Kramer, private communication
(59) M. Soell, private communication, 1975.
(63) B. S. Brown, J. W. Hafstrom, and T. Klippert, to be published.
(68) S. Colucci, H. Weinstock, and B. S. Brown, to be published.
(73) C. L. Snead Jr. and D. M. Parkin, private communication.


(83) E. Seibt, private communication.

