The Effect of Irradiation Temperature on REBCO $J_c$
Degradation and Implications for Fusion Magnets

by
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Abstract

Recent advances in high temperature superconductors (HTS) have opened up a new parameter space for the design of tokamak fusion pilot plants. While previously the maximum on-axis field in a superconducting tokamak was limited to ~6 T, HTS allows tokamaks to be designed with much higher on-axis fields, leading to smaller reactor designs. For these designs, it is critical to determine the lifetime of modern HTS technology in an environment relevant to compact, high-field fusion reactors as well as develop strategies to mitigate this damage. While some studies have been undertaken to assess the lifetime of coated conductors in a fast neutron environment, facilities do not exist to perform cryogenic neutron irradiations at the present. In addition, reactor studies are costly and activate the samples, requiring long cooldown times and specialized analysis facilities to handle radioactive material. In order to complement reactor irradiation studies of HTS and determine whether elevated temperature irradiation has an effect on $J_c$ degradation, REBCO coated conductors were irradiated with a 1.2 MeV proton beam at 80 K, 323 K, and 423 K. Proton irradiation at cryogenic temperatures was found to substantially reduce the amount of $J_c$ degradation in the REBCO samples irradiated to high fluences, a result of great importance to superconducting REBCO magnets in fusion applications where the radiation will occur at $T < 80$ K. An analysis of temperature, field, and angle dependencies of $J_c$ was performed to investigate the microstructural mechanisms behind the $J_c$ degradation at different temperatures. The key mechanism driving the differences in $J_c$ degradation was found to be radiation-enhanced diffusion at higher temperatures, leading to grain boundary widening between superconducting crystals which in turn blocked supercurrent transport through the conductor. Molecular dynamics simulations suggest that the same mechanism (enhanced diffusion to grain boundaries) also applies to neutron irradiations. This motivates a re-evaluation of previous REBCO neutron irradiation studies at temperatures between 323 K and 383 K, specifically with regards to predictions about REBCO lifetimes in a fusion environment. The work in this thesis suggests that at cryogenic temperatures, the $J_c$ degradation observed in these studies could be substantially less than previously reported.
Thesis Supervisor: Dennis G. Whyte
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Thesis Reader: Michael Short
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I am a bit lost again - as you can tell, I get lost all the time. How can technology that will be available in 40 to 80 years possibly influence climate change?
— Lord Peston, UK Fusion Review, 2015

Superconductor is almost too wonderful. You end up using it in everything.
— Larry Niven, Ringworld

In this review, over 200 HTS-related papers were scanned... Due to the variety of samples and their quality, irradiation beams, irradiation temperature, and different measurement methods, some results are confusing and sometimes conflicting.
— Wei-Kan Chu, Review Paper of YBCO Irradiation Damage Studies
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When most people think of scientific research, the image comes to mind of a lone researcher burning the midnight oil in the lab. Nothing could be further from the truth. Behind every successful PhD project is an army of mentors, collaborators, and friends, and nowhere is that more true than here at MIT. A few pages is nowhere near long enough to express my gratitude for your support and friendship, but I’ll give it my best shot.

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4.3.1 Repeatability of measurements and error analysis 80

5 Critical Current Analysis of Radiation Damaged REBCO 85
5.1 Critical temperature modifications 87
5.2 Differentiating strong and weak pinning regions 89
5.3 $J_c$ vs. $\theta$ comparisons 92
5.4 $J_c$ vs. B comparisons 95
5.5 Grain boundary-dominated vs. pinning-dominated field regions 97
5.6 Effect of annealing 100

6 Molecular Dynamics Modeling of REBCO 101
6.1 Assumptions and limitations of the simulations 102
6.2 Simulation workflow 102
6.2.1 Monte Carlo determination of incident irradiation energies 103
6.2.2 Binary collision approximations of PKA energy spectra 104
6.2.3 Molecular dynamics simulations 106
6.3 Defect formation in irradiated YBCO 109
6.3.1 Simulation setup 109
6.3.2 Defect formation comparisons 111
6.4 Oxygen diffusion in YBCO 113
6.4.1 Mean-square-displacement simulations 114
6.4.2 Calculation of diffusion coefficients 116
6.5 Applicability of REBCO ion irradiation results to fusion neutron irradiation 118
6.5.1 Defect formation 120
6.5.2 Gadolinium effects 122

7 Conclusions and Recommendations for Future Work 125
7.1 Major accomplishments of this thesis 126
7.2 Implications for future devices 126
7.3 Suggestions for future work 127
7.3.1 Cryogenic, in-core reactor irradiation 128
7.3.2 High-fidelity analysis of neutron-irradiated samples 128
7.3.3 In-situ accelerator $I_c$ analysis 128
7.3.4 Further annealing studies 128
7.3.5 TEM comparisons of ion and neutron irradiated tapes 129
7.3.6 Isolating thermal neutron effects on different tape chemistries 129

A Detailed DANTE Repair Information 131
A.1 Inductor design 131
A.2 Normal operating conditions 132

B Molecular Dynamics Simulations 137
B.1 LAMMPS input files 137
B.1.1 MSD input file 137
## List of Figures

<table>
<thead>
<tr>
<th>Figure</th>
<th>Description</th>
<th>Page</th>
</tr>
</thead>
<tbody>
<tr>
<td>1-1</td>
<td>The ARC reactor, shown with the plasma in yellow and the TF superconducting tape in brown. Note the neutron shield is omitted for viewing clarity. Also note that although the ARC design is based on a diverted plasma, the physical divertor design was left for later study and a simplified representation of the vacuum vessel is shown here.</td>
<td>27</td>
</tr>
<tr>
<td>1-2</td>
<td>The ARC reactor inboard radial build.</td>
<td>28</td>
</tr>
<tr>
<td>1-3</td>
<td>Scale comparison of ITER and ARC.</td>
<td>28</td>
</tr>
<tr>
<td>1-4</td>
<td>The upper half of ARC's superconducting coils can be removed, allowing the vacuum vessel to be removed from the blanket tank as a single piece.</td>
<td>29</td>
</tr>
<tr>
<td>1-5</td>
<td>Cross section of the axisymmetric geometry used to model neutron transport in ARC using MCNP. Yellow indicates the plasma region, orange indicates the TiH₂ shielding, light blue indicates FLiBe, and brown indicates the superconducting coil structure. Gray indicates either Inconel 718 or tungsten for the vacuum vessel, connection post, and divertor. The “squaring” of some corners in this simplified model has a negligible effect on the neutronics analysis.</td>
<td>31</td>
</tr>
<tr>
<td>1-6</td>
<td>Cutaway of results of ARC MCNP neutronics simulation, showing where the model in Figure 1-5 corresponds to the reactor concept. Note the increased flux at the inner midplane, which is the limiting factor for coil lifetime and indicative of the neutronics challenges that compact, high-field fusion reactors face.</td>
<td>32</td>
</tr>
<tr>
<td>1-7</td>
<td>Comparison of ARC neutron energy spectrum at the magnet coils with the in-core MIT Reactor fission neutron spectrum. The flux present at lower neutron energies for the ARC spectrum is due to the down-scattering of neutrons by the blanket and shield before reaching the magnet coil.</td>
<td>33</td>
</tr>
<tr>
<td>2-1</td>
<td>Schematic of a critical surface of a superconductor [42]. The shape of the surface is caused by the dependences of ( J_c ), ( B_c ) and ( T_c ) on each other and is different for each type of superconductor.</td>
<td>36</td>
</tr>
</tbody>
</table>
2-2 Schematics of free energy density contributions from electron ordering (negative contribution with a length scale of $\xi$) and magnetization (positive contribution with a length scale of $\lambda$) [48]. The sum of the two contributions determines whether the boundary between superconducting and normal phases is thermodynamically stable, expelling all magnetic flux (Type I superconductor) or unstable, allowing some flux to penetrate (Type II superconductor).

2-3 SQUID microscopy image of flux vortices in YBCO [54]. It is important to note that lack of resolution on the SQUID microscope is responsible for the larger-than-expected vortex diameters in this image. Nevertheless, the centers of the vortices should be accurate and thus the distribution (i.e. approximately triangular lattice) of the flux vortices is experimentally observed.

2-4 Schematic of normal core and vortex dimensions in an anisotropic superconductor such as REBCO where $\xi$ and $\lambda$ have different values depending on which crystalographic axis is being investigated [55]. Normal cores are characterized by $\xi$ and vortex current loops are characterized by $\lambda$.

2-5 Orthorhombic structure of the REBCO unit cell, generated with the VESTA 3D visualization program [60] on the left, along with a repeated structure to illustrate the three different superconducting planes (each of which repeats once in the c-direction per unit cell) on the right. Oxygen is red, copper is blue, barium is green, and the rare earth element is gray. The highly anisotropic structure of REBCO contributes to the differences in critical current at different applied field angles.

2-6 Comparison of LTS (red) and HTS (green) critical surfaces, demonstrating the order-of-magnitude increase in all three parameters in HTS [courtesy of D. Brunner].

2-7 Schematic view of the cross section of REBCO tape superconductor by SuperPower Inc. Each layer is shown with its typical thickness. The tape is normally available in widths between 2 and 12 mm, and a total thickness of between 0.05 to 0.1 mm [courtesy of F. Mangiarotti].

2-8 Schematic of normal core formation and pinning site force contributions which counteract the Lorentz force on the flux lines when current is passed through the superconductor.

2-9 Schematics of strong and weak pinning sites. The strong pinning site deforms the shape of the flux line, while the weak pinning sites act collectively to pin a flux line and it retains its shape.

2-10 $J_c$ vs. $\theta$ plot showing correlated pinning at 0, 90, and 180 degrees to the $ab$ plane. The 90 degree peak arises due to the intrinsic Cu-O chain layer pinning and the 0 and 180 degree peak arises due to the 1D BZO nanorod artificial pinning sites oriented along the $c$-axis of the superconductor.
2-11 TEM images showing examples of 1D, 2D, and 3D pinning sites (0D sites are too small to be resolved with a TEM): 2-11a BZO nanorods, 2-11b Cu-O chain layers, and 2-11c 3D precipitates. Examples of pinning sites are pointed out or shaded in blue to guide the eye.

2-12 Normalized critical temperature dependence on hole concentration for a wide variety of cuprate superconductors [72]

2-13 TEM image of a 30 degree [001] tilt grain boundary in YBCO. The structural units of the grain boundary have been highlighted in color to show the approximate width (~1 nm) of the grain boundary [87].

3-1 Schematic of the DANTE accelerator, with major subsystems annotated [90].

3-2 Schematic of DANTE negative ion source [90].

3-3 Multicusp configuration in negative ion source. The mirror effect produced by the magnetic field lines (in red) confines the source plasma (yellow).

3-4 DANTE accelerator with column removed for inspection. The low-energy section is to the left of the terminal and the high-energy section is to the right of the terminal.

3-5 Beam emittances and spatial profile for the ion source optics. The negative hydrogen ions (red) are accelerated through the ion source while the electrons (yellow) are steered into the extraction electrode with a set of filtering magnets. The ion trajectories are slightly affected by the filter magnets (as seen by the offset in emittance plots from center) but this is corrected later in the Einzel lens.

3-6 Visible discoloration on extraction electrode corresponds to simulation results of diverted electron beam.

3-7 Beam simulation through the whole Einzel lens. Note how the beam center has been moved almost entirely back to the center of the ion source by the focusing of the Einzel lens. Emittance and YZ beam profile plots taken at the exit of the Einzel lens.

3-8 The filament is connected to two leads, insulated from ground, and the gas feedthrough is visible below the filament leads.

3-9 DANTE energy calibration data and fit.

3-10 SRIM simulations for 1.2 MeV protons on silver-capped REBCO tape. These simulations show that for the samples being investigated, the beam particles are deposited far beneath the REBCO layer and the average energy to recoils in the REBCO layer is approximately constant.

3-11 Comparison of primary recoil energy spectra for 800 keV, 1 MeV, and 1.2 MeV proton beams on YBCO to illustrate the insensitivity of primary recoils to incident proton energy within a few hundred keV of the average proton energy 1 MeV in the REBCO. The spectra are overlaid for most of the energy range and only diverge at energies close to the beam energies themselves.

3-12 BEATRICE image analysis of a typical beamspot.
3-13 The beam image in 3-12 was scaled from pixels to mm and vertical and horizontal slices of the beam profile were obtained. In each slice, a second scale was placed at 75% of the peak intensity in order to obtain an estimate of the beam width at 75% of full intensity. Note that the beam has been tuned so that the Gaussian peak is not symmetric in 2D, as the 6x4 mm collimator is wider in the horizontal direction.

3-14 Image of HTS target mount. In Figure (a), the top disk is the first collimator, followed by the secondary electron suppression electrode, with the secondary G-10 collimator mount at the bottom. Figure (b) is a close-up of the target area with the collimator and suppression electrode removed, showing the four current pickups used to center the beam on target during operation.

3-15 Comsol target heating model displaying maximum target temperature rise (at steady state) for a 1 μA at 1 MeV beam heat load with a starting temperature of 300 K.

3-16 HTS cryogenic irradiation chamber.

3-17 Traces of target temperature (a) and chamber pressure (b) vs. time for one of the cryogenic irradiations. Current is also plotted in both figures to indicate when the beam is on target. Although the beam current changes due to changing plasma conditions in the ion source, the target temperature and pressure are maintained at approximately constant levels throughout the irradiation.

4-1 Schematic of four-probe critical current measurement. Orange represents the superconducting REBCO tape, while the other components (gray and black) are normal conductors. A power supply is connected to both ends of the superconductor (while it is cold) to provide current. Voltage is measured across a known distance as current is increased and an electric field criterion (typically a rise of 1 μV/cm) is used to determine the transition between the superconducting and normal state.

4-2 Example $I_c$ transport measurement for a sample with 0.4 cm between voltage taps. Note that the fit bounds have been adjusted to exclude the noisy baseline region near zero current as well as the region at high current where the superconductor starts to become fully normal. The n-value for this measurement was 18.7.

4-3 SuperCurrent automated $I_c$ measurement system (left) with control/power supply cabinet (right).

4-4 Example of a normal (undoped) REBCO critical current angular dependence, with a peak at 90 degrees and a minimum close to 0 and 180 degrees.

4-5 Example of angular dependence caused by artificial correlated pinning (in this case due to BZO nanorod inclusions). Note that the 0 and 180 degree currents (typically the minimum current) are almost as high as the 90 degree current.
4-6 Sample removal from the G10 irradiation stage coupon. Care was taken to not bend or scratch the samples during removal, which is why only plastic and wooden tools were used for this process. 78

4-7 Sample preparation process. Figure a shows the sample after the application of the photolithography mask. Figure b shows the sample after silver etching (the exposed YBCO layer is black). Figure c shows the sample after the YBCO etching (the exposed buffer layer is a reflective green color). The final bridge pattern is shown in Figure d. 79

4-8 Sample mounted before insertion in the SuperCurrent system with along with a schematic showing the location of the current leads and voltage tapes. 80

4-9 SuperCurrent control and monitoring software. 81

4-10 Determination of SuperCurrent measurement repeatability. Each data-point represents the relative standard deviation for three repeat I_c measurements taken at the same temperature, field, and field angle. The average relative standard deviation is 1.3% of the measured critical current value. 82

4-11 TapeStar critical current measurements used to normalize baseline I_c values for irradiated samples. 82

4-12 Comparison of critical current measurements between a normal beam flux and 3x reduced beam flux at a given fluence. The critical current measurements are similar, leading to the conclusion that dose rate, at the damage rates used in this study, does not play a large role in I_c degradation due to irradiation. 83

4-13 Comparison of critical current measurements between an unirradiated sample heated to 423 K for 80 minutes and the unirradiated, unheated control. An observation of the two plots shows that sample heating by itself does not lead to significant I_c degradation at angles besides 90 degrees. 84

5-1 Critical current density (at 5 T, 30 K) of samples irradiated at different temperatures to fluences of \(1 \times 10^{16} \text{ p/cm}^2\) and \(5 \times 10^{16} \text{ p/cm}^2\). Lines are drawn between data points to guide the eye. \(J_c\) degradation shows a clear dependence on irradiation temperature at the higher fluence. 85

5-2 Selection of plots illustrating irradiation temperature effect on REBCO \(J_c\) degradation due to proton irradiation at various measurement fields and temperatures, with lines connecting data points to guide the eye. There appears a transition between \(1 \times 10^{16} \text{ p/cm}^2\) and \(5 \times 10^{16} \text{ p/cm}^2\) from a regime which is minimally affected by irradiation temperature to a regime in which there is a large difference between cryogenic and high temperature irradiation conditions. 86

5-3 Zero-field \(T_c\) fit on the pristine control sample. 88

5-4 Critical temperature vs. irradiation temperature. Note that the zero of the y axis has been suppressed to highlight the changes between fluences. 88
5-5 Critical temperature vs. irradiation temperature for a fluence of $1 \times 10^{16}$ p/cm$^2$. The annealed sample recovered to approximate the same critical temperature as both higher-temperature irradiations.

5-6 Comparison of measured $J_c$ to fit with temperature in Eq. 5.1 (dotted line). Vertical dashed lines indicate the approximate transition temperature between weak and strong pinning regimes.

5-7 Comparison of measured $J_c$ to fit with temperature in Eq. 5.1 (dotted line). Vertical dashed lines indicate the approximate transition temperature between weak and strong pinning regimes.

5-8 Comparison of measured $J_c$ to fit with temperature in Eq. 5.1 (dotted line). The region where the log($J_c$) data is linear with temperature (thus matching the dotted fit line) is where weak pinning is active and the region where the data diverges from the fit indicates a transition from weak to strong pinning.

5-9 Critical current density vs. measurement angle for 80 K, $5 \times 10^{15}$ p/cm$^2$ irradiated sample as compared to the unirradiated sample, in both strong (left) and weak (right) pinning regimes.

5-10 Critical current density vs. measurement angle for 80 K, $1 \times 10^{16}$ p/cm$^2$ irradiated sample as compared to the unirradiated sample, in both strong (left) and weak (right) pinning regimes.

5-11 Critical current density vs. measurement angle for 80 K, $5 \times 10^{16}$ p/cm$^2$ irradiated sample as compared to the unirradiated sample, in both strong (left) and weak (right) pinning regimes.

5-12 Critical current density vs. measurement angle for 423 K, $5 \times 10^{15}$ p/cm$^2$ irradiated sample as compared to the unirradiated sample, in both strong (left) and weak (right) pinning regimes.

5-13 Critical current density vs. measurement angle for 423 K, $1 \times 10^{16}$ p/cm$^2$ irradiated sample as compared to the unirradiated sample, in both strong (left) and weak (right) pinning regimes.

5-14 Critical current density vs. measurement angle for 423 K, $5 \times 10^{16}$ p/cm$^2$ irradiated sample as compared to the unirradiated sample, in both strong (left) and weak (right) pinning regimes.

5-15 $J_c$ vs. $B$ for control sample with fitted power law $\alpha$ values. Error is represented as a shaded region.

5-16 $J_c$ vs. $B$ curves for 80K irradiations with calculated power law $\alpha$ values. Error is represented as a shaded region.

5-17 $J_c$ vs. $B$ curves with for 423K irradiations with fitted power law $\alpha$ values. Error is represented as a shaded region.
Crossover between grain-boundary limited \( J_c \) and pinning limited \( J_c \) regimes for 80 K irradiations. Error is represented as the shaded region and are connected between data points to guide the eye to where the crossover occurs. At fluences higher than \( 5 \times 10^{15} \) p/cm\(^2\), a decrease in the irradiated \( J_c \) compared to the pristine sample is observed, and the crossover field shifts to higher values of B at higher fluences, reflecting increasing grain boundary disorder becoming dominant over the pinning contributions of the defects at higher field regions.

Crossover between grain-boundary limited \( J_c \) and pinning limited \( J_c \) regimes for 80 K and 423 K irradiations at a fluence of \( 5 \times 10^{16} \) p/cm\(^2\). Error is represented as the shaded region and are connected between data points to guide the eye to where the crossover occurs. The higher temperature irradiation has a crossover point outside of the achievable measurement range (> 8T), indicating significant grain boundary widening at the higher temperature irradiation.

Angular dependence of annealed vs. un-annealed samples irradiated to \( 1 \times 10^{16} \) p/cm\(^2\) at 80 K.

Simulation workflow, illustrating inputs and outputs of the analysis codes used in this thesis.

Comparison of PKA energy cumulative distribution functions resulting from 1 MeV protons and fusion neutrons, generated by the DART code. The large difference in PKA energies for the two types of primary irradiation is due to the different interaction mechanisms governing the collisions (Coulomb scattering vs. nuclear scattering) and the large range of PKA energies from neutron irradiation are due to the wide range of neutron energies incident at the TF coil.

YBCO volume modeled in LAMMPS.

Progression of cascade in YBCO, for Ba [001] PKA of energy 10 keV at a simulation temperature of 423 K. Sites are color coded according to atom type (i.e. Cu is blue, O is red, Ba is green, and Y is gray) and represent either vacancy or interstitials. After approximately 20 ps the defect cluster has fully quenched from the cascade.

Comparison of 80 K and 423 K baseline MD simulations.

Comparison of 80 K 50 eV and 5 keV MD simulations. Red particles indicate interstitial sites and blue particles indicate vacancy sites.

Comparison of vacancy (left) vs. interstitial (right) sites for the Ba [001] 5 keV PKA simulation in Figure 6-6.

Comparison of 80 K and 423 K defect clusters produced by a 5 keV, [001] directed Ba PKA in YBCO. Red particles indicate interstitial sites and blue particles indicate vacancy sites.

Comparison of 80 K and 423 K 10 keV MD simulations. Red particles indicate interstitial sites and blue particles indicate vacancy sites. The defects at the top of the simulation are result of periodic boundary conditions.
6-10 Comparison of 80 K and 423 K radial distribution functions for clusters produced by 5 keV [001] Ba PKAs. The smoothing of peaks for the 423 K g(r) indicates the amorphous nature of the defect. 116

6-11 Frenkel pair production (per PKA) vs. PKA energy for 80K and 423 K 117

6-12 800 K MSD results and fit to determine diffusion coefficient. 118

6-13 Arrhenius fit to 700, 800, 900, and 1000 K diffusion coefficients determined from MSD simulations. 119

6-14 Frenkel pair production (per PKA) vs. PKA energy for O [001], [010], and [100] directed PKAs at 20K. 120

6-15 Frenkel pair production (per PKA) vs. PKA energy for Ba and O [001] PKAs at 20K. 121

6-16 Frenkel pair production per PKA vs. PKA energy for O [001] PKAs at 20K, 300K, and 423K. 122

A-1 Calculation of required dimensions and turns for replacement 1 H inductor in DANTE. 131

A-2 CAD drawing of the former used to wind the inductor. 132

A-3 Final installation of custom-built inductor. 134

A-4 DANTE control screen. 134

A-5 Magnet power supplies providing current to the selection (top), horizontal (second down) and vertical (third down) steering magnets, and focusing quadrupoles (bottom two supplies). 135

A-6 Typical trace of terminal switching power supply signal. 135
List of Tables

1.1 List of significant ARC design parameters. ........................................ 26

2.1 Coherence lengths in YBCO for different temperatures and orienta-
tions. The subscripts indicate the directionality of the coherence length
with respect to the crystallographic axes of the superconductor. ............. 37

3.1 Optimal DANTE beam parameters .................................................. 62

3.2 Location and width of $^{19}$F($p$, $\gamma\alpha)^{16}$O resonance gamma peaks used to
calibrate DANTE beam energy. .......................................................... 65

6.1 Coefficients used in the Chaplot potential. ........................................ 107

6.2 Effective atomic charges and radii used in the Chaplot potential for $\delta$
$= 0$. ................................................................................................. 108

6.3 Baseline Frenkel pair production for the entire simulation volume. ... 110

6.4 Diffusion Coefficient Extrapolations ................................................. 117

6.5 Diffusion coefficient extrapolations to temperatures relevant to neutron
irradiation studies as well as fusion conditions. ................................. 122

A.1 List of significant DANTE operational parameters, recorded during
March 29, 2017 irradiations. Nominal beam current on target was
$\sim 300$ nA and could be increased/decreased by adjusting the filament
current. Note that values quoted are the actual values (represented in
green text in Figure A-4), not the set point values. ............................... 133
Chapter 1

Introduction

Fusion energy has been long-sought as the solution to the growing global demand for energy. Fusion reactors would burn isotopes of hydrogen to produce helium, and reserves of the basic fuel (deuterium and lithium to breed tritium) are essentially limitless, providing a sustainable source of energy for the foreseeable future. While activation of the structural fusion reactor materials will create a small amount of short-lived nuclear waste, fusion reactors would produce far less (and far less potent) radioactive waste compared to conventional fission reactors. Like fission, fusion reactors would produce no harmful particulate or greenhouse emissions, allowing fusion reactors to be part of a “clean” energy mix required to combat climate change. Unlike other clean energy sources such as wind and solar however, fusion reactors would be able to operate continuously, providing baseload electricity [1].

A fusion reactor would operate by containing a thermonuclear plasma within some kind of confinement field. While stars are large enough to use gravity to confine their thermonuclear plasma, a terrestrial fusion reactor is too small for this confinement method and must rely on either electric or magnetic fields to confine the fusion plasma\(^1\). Although many electromagnetic configurations have been tested in the past 50 years, the leading candidate in fusion performance has been a donut-shaped magnetic confinement configuration called a tokamak. While progress in tokamak fusion confinement has steadily increased, device sizes have also increased, leading to the current flagship fusion experiment ITER [2] with costs in the tens of billions of dollars and a construction time of decades. It is not clear that proposed demonstration power plants [3] (which would be even larger than ITER) could be built on shorter timescales or less cost, calling into question the current embodiment of fusion reactors as a clean energy solution to tackle the shorter-term challenges of global energy demand and climate change.

In order for magnetic fusion to be developed on a reasonable timescale and provide

\[^1\]Pulsed fusion reactions can be achieved using a high-energy driver, such as a fission explosion or very large array of lasers, to rapidly compress the fusion fuel, but this thesis will only focus on steady-state fusion reactions
an economically competitive, and therefore useful, energy source it is necessary to minimize the fusion core size at a given fusion power output. When high-temperature superconductors (HTS) are applied to high-field tokamak fusion power plant design, it is no longer the plasma confinement performance that is the limiting factor to shrinking the device size; sufficient confinement can be obtained by increasing the magnetic field strength. Instead, the largest constraint in reducing the size was found to be lifetime radiation damage limits to the HTS coils due to reduced room for inboard shielding of the TF coils from the high-energy neutrons from the core [4]. This is in stark contrast to larger reactor designs such as EU demo [3], which have ample room for coil shielding and have so far driven all fusion-related REBCO damage studies to date [5, 6]. If compact, high-field tokamak designs are to be pursued, the lifetime of the TF coils will be a key economic driver, and it is thus imperative to determine the absolute limits of HTS operation in a fast neutron environment as well as develop strategies to mitigate this damage.

1.1 High field approach to fusion

Although ceramic-based HTS were first discovered in 1987, large scale production of rare-earth barium copper oxide (REBCO)\(^2\) HTS conductor was not possible until recently due to the difficulty in manufacturing long strands of REBCO which still retain high performance. Advances in deposition precursor methods such as RABiTS and IBAD [7, 8] have allowed the production of kilometer-length strands of REBCO in the past few years, opening up the possibility of using HTS to wind large bore magnets for use in fusion devices. Previously, the maximum on-coil field in a superconducting tokamak was limited to approximately 13 T [9], constraining the on-axis field to \(~6\) T for a standard aspect ratio \((R/a \sim 3)\). However, the expanded operational space in field, current, and temperature of REBCO removes this constraint. The lack of significant critical current degradation of REBCO (discussed in Chapter 2) at high magnetic fields allows tokamaks to be designed with much higher on-axis fields. Access to higher fields significantly relaxes plasma physics constraints and allows smaller devices at higher fields to access the same performance as larger devices with lower fields. The ARC reactor design study [4] showed that the use of REBCO to design a compact, high-field pilot plant enabled a 3.3 m major radius device to achieve a fusion power of 525 MW, comparable to the performance of ITER [2].

\(^2\)It is important to note that the earliest and most commonly studied form of REBCO was YBCO (using yttrium as the rare earth element). Thus, much of the physics basis for the understanding of REBCO was based on experiments with YBCO. As HTS became commercialized, manufacturers began substituting other rare earth elements such as gadolinium for yttrium. In some cases, the “RE” component is some combination of several rare earth elements, the exact composition being proprietary to the conductor manufacturers. Since most of the radiation damage effects discussed in this thesis deal mainly with oxygen and copper displacement effects, the general term REBCO will be used to refer to the rare-earth cuprate superconductor family.
1.2 The ARC reactor

Most fusion reactor designs, such as the ARIES studies [10, 11, 12, 13], assume a large, fixed 1000 MWe output for a power plant. However, large-scale designs make fusion engineering research and development difficult because of the high cost and long construction time of experiments. In order to relax this size constraint and incorporate the advantages of a high-field design utilizing REBCO superconducting magnets, the 200 MWe ARC reactor was designed [4]. The design was carried out as a follow-on to the Vulcan conceptual PMI device design which also utilized demountable REBCO tape [14]. An explicit design goal of ARC was to minimize the reactor size in order to reduce the plant capital cost. Like Vulcan, ARC makes use of high-temperature superconductors (HTS), which enables large on-axis magnetic fields and ultimately reduces the size of the reactor.

The ARC design process proceeded as follows: first a 0-D point design was carried out using simple scaling laws to establish baseline values for volume-averaged quantities and basic reactor dimensions. After the 0-D design study was performed, a 2-D plasma core analysis was performed with the ACCOMNE code [15] using scaled experimental profiles. At the same time, the MCNP5 neutron transport code [16] was used to assess the neutronics performance of a 2-D axisymmetric reactor geometry and the COMSOL multiphysics package [17] was used to assess 2-D and 3-D models of the reactor structure and liquid blanket thermal performance. The design was iterated through these three codes until an acceptable set of reactor parameters was achieved.

The reactor design is shown in Fig. 1-1, the inboard radial build in Fig. 1-2, and the most significant design parameters are given in Table 1.1. The starting objective of the ARC study was to determine if a reduced size D-T fusion device (fusion power ≤ 500 MW) could benefit from the high magnetic field technology offered by recently developed high temperature superconductors. The reasoning was that a high magnitude magnetic field in a compact, superconducting device might offer not only access to high plasma gain $Q_p$, but also enable net electric gain $Q_e > 1$. This specific option has not been explored previously in design studies, although the recent advanced tokamak (AT) Pilot ($Q_e=1$) study of Menard et al. [18] had similar design goals, but used conventional superconductor technology. A recent FNSF study is the FDF design [19], which is a similar size to ARC, but consumes > 500 MW of electricity because it does not use superconducting magnets. As an illustration of the reduction of reactor size achieved, the designed fusion power of ARC is slightly higher than the designed fusion power of ITER, which has $R = 6.2$ meters (comparison shown in Figure 1-3.

The use of REBCO superconducting technology in the toroidal field (TF) coils permits significantly higher on-axis magnetic fields than standard Nb$_3$Sn superconductors. High magnetic field strength is essential in small reactor designs in order to achieve the necessary poloidal field/plasma current needed for sufficient confinement and stability against beta (pressure) limits. In addition, when holding beta constant the volumetric
<table>
<thead>
<tr>
<th>Design Parameter</th>
<th>Symbol</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Fusion power</td>
<td>$P_f$</td>
<td>525 MW</td>
</tr>
<tr>
<td>Total thermal power</td>
<td>$P_{tot}$</td>
<td>708 MW</td>
</tr>
<tr>
<td>Plant thermal efficiency</td>
<td>$\eta_{elec}$</td>
<td>0.40</td>
</tr>
<tr>
<td>Total electric power</td>
<td>$P_e$</td>
<td>283 MW</td>
</tr>
<tr>
<td>Net electric power</td>
<td>$P_{net}$</td>
<td>190 MW</td>
</tr>
<tr>
<td>LHCD coupled power</td>
<td>$P_{LH}$</td>
<td>25 MW</td>
</tr>
<tr>
<td>ICRF coupled power</td>
<td>$P_{IC}$</td>
<td>13.6 MW</td>
</tr>
<tr>
<td>Power multiplication factor</td>
<td>$Q_e$</td>
<td>3.0</td>
</tr>
<tr>
<td>Major radius</td>
<td>$R_0$</td>
<td>3.3 m</td>
</tr>
<tr>
<td>Plasma semi-minor radius</td>
<td>$a$</td>
<td>1.13 m</td>
</tr>
<tr>
<td>Plasma elongation</td>
<td>$\kappa$</td>
<td>1.84</td>
</tr>
<tr>
<td>Plasma volume</td>
<td>$V_p$</td>
<td>$141 \text{ m}^3$</td>
</tr>
<tr>
<td>Toroidal magnetic field</td>
<td>$B_0$</td>
<td>9.2 T</td>
</tr>
<tr>
<td>Peak on-coil magnetic field</td>
<td>$B_{max}$</td>
<td>23 T</td>
</tr>
<tr>
<td>Plasma current</td>
<td>$I_p$</td>
<td>7.8 MA</td>
</tr>
<tr>
<td>Bootstrap fraction</td>
<td>$f_{BS}$</td>
<td>0.63</td>
</tr>
<tr>
<td>Tritium Breeding Ratio</td>
<td>TBR</td>
<td>1.1</td>
</tr>
<tr>
<td>Avg. temperature</td>
<td>$\langle T \rangle$</td>
<td>14 keV</td>
</tr>
<tr>
<td>Avg. density</td>
<td>$\langle n \rangle$</td>
<td>$1.3 \times 10^{20} \text{ m}^{-3}$</td>
</tr>
<tr>
<td>On-axis temperature</td>
<td>$T_0$</td>
<td>27 keV</td>
</tr>
<tr>
<td>On-axis density</td>
<td>$n_0$</td>
<td>$1.8 \times 10^{20} \text{ m}^{-3}$</td>
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<tr>
<td>Greenwald fraction</td>
<td>$f_{Gr}$</td>
<td>0.67</td>
</tr>
<tr>
<td>Toroidal beta</td>
<td>$\beta_T$</td>
<td>1.9%</td>
</tr>
<tr>
<td>Internal inductance</td>
<td>$l_i$</td>
<td>0.67</td>
</tr>
<tr>
<td>Normalized beta</td>
<td>$\beta_N$</td>
<td>2.59</td>
</tr>
<tr>
<td>Safety factor at $r/a = 0.95$</td>
<td>$q_{95}$</td>
<td>7.2</td>
</tr>
<tr>
<td>Edge safety factor</td>
<td>$q_a$</td>
<td>4.7</td>
</tr>
<tr>
<td>Minimum safety factor</td>
<td>$q_{min}$</td>
<td>3.5</td>
</tr>
<tr>
<td>Fusion power wall loading</td>
<td>$P_f/S_b$</td>
<td>2.5 MW/m²</td>
</tr>
<tr>
<td>Energy confinement time</td>
<td>$\tau_E$</td>
<td>0.64 sec</td>
</tr>
<tr>
<td>H89 confinement factor</td>
<td>$H_{89}$</td>
<td>2.8</td>
</tr>
<tr>
<td>H98(y,2) confinement factor</td>
<td>$H_{98y2}$</td>
<td>1.8</td>
</tr>
<tr>
<td>G89 gain factor</td>
<td>$G_{89}$</td>
<td>0.14</td>
</tr>
</tbody>
</table>

Table 1.1: List of significant ARC design parameters.
fusion power scales as $\sim B_0^4$ and, at constant safety factor, the plasma confinement strongly improves with magnetic field strength [20]. Another main advantage of using REBCO is that it enables resistive joints in the superconducting coils [21], and thus the TF coils can be made demountable, meaning the coils can be split into two pieces (see Fig. 1-4). As discussed below, demountability can provide a dramatically different and likely more attractive, modular maintenance scheme for magnetic fusion devices. The trade-offs between modular component replacement [22] and power dissipation in the TF joints [21] have only been explored at small size in the Vulcan D-D device ($R_0 \sim 1.2$ m), which motivates our exploration of demountability in a D-T reactor.

With non-demountable TF coils, it is necessary to split all components located inside the TF coils into toroidal sections that can fit between the gaps in the TF coils (referred to as sector maintenance), which is complex and time-intensive. Sector maintenance necessitates significantly larger TF coils to allow for space to remove
sections of the vacuum vessel (e.g. ARIES-AT [11]). With joints, the TF coils, which are the most expensive component of a reactor [23], can be made smaller. When coupled with a liquid immersion molten salt blanket and a compact, high-field design, the vertical maintenance scheme enabled by demountable toroidal field coils would allow simplified removal and replacement of the entire vacuum vessel as a single component. The vacuum vessel module could be constructed and tested entirely offsite and be relatively quickly lowered into place with the TF coils demounted, minimizing or eliminating the maintenance that must be performed inside the TF volume itself. The relative ease of installation and external testing of all internal components should greatly increase the simplicity and reliability of component replacement.

Demountability is a particularly attractive feature for an FNSF and was the motivation for using demountable copper coils in the FDF design [19]. Demountable vacuum vessels would allow early fusion reactors to simultaneously perform as fusion nuclear science facilities (FNSFs) by effectively moving the safety barrier out from the vacuum vessel to the blanket tank and using each replacement of the vacuum vessel as the opportunity to test new nuclear materials. This breaks out of the “chicken and egg” problem of having to design an FNSF vacuum vessel with materials which are known beforehand to be robust to the nuclear environment in a fusion device. Demountable
Figure 1-4: The upper half of ARC's superconducting coils can be removed, allowing the vacuum vessel to be removed from the blanket tank as a single piece.

copper TF coils have already been used in experimental devices such as COMPASS [24], DIII-D [25] and Alcator C-Mod [26].

Looking ahead to commercial power plants, a vertical maintenance scheme as described above would have substantially faster maintenance times than a device with sector maintenance, allowing the availability of the fusion reactor to be higher. Plant availability is known to be a key driver for fission reactor economics [27], and this would be the case with fusion reactors as well. Increased availability would greatly improve the economic viability of fusion reactors, leading to a higher chance of fusion being adopted as a source of clean power.

1.2.1 Neutronics challenges unique to high field approach

Superconducting magnets of all types have been shown to be sensitive to high-energy neutron radiation [5]. However, all fusion-relevant HTS irradiation studies to date have been predicated on the existence of a large (i.e. major radius R > 6 m) reactor which will have ample shielding allowed in the radial build. Another important difference between HTS magnets and LTS magnets is that LTS magnets require a large amount of electrical insulation which is typically more sensitive to radiation than the superconductor itself [28]. These two factors have driven fusion irradiation studies of HTS (specifically REBCO) for fusion applications in the past two decades. Due to the high cost and difficulty of handling radioactive samples from fission reactor irradiations, neutron irradiation studies to date have understandably only investigated
the effects of radiation damage up to a certain amount which is “good enough” to qualify coated conductors for use in a large fusion reactor. This fluence is generally understood to be approximately $3 \times 10^{18}$ neutrons/cm$^2$ of “fast” neutrons with energy $> 0.1$ MeV [29]. In a large device, such as ITER, fluence to the coils is only expected to be $\sim 2 \times 10^{18}$ neutrons/cm$^2$ over the lifetime of the device.

In compact, high-field reactors utilizing HTS, this situation changes significantly. As high-field magnets allow small, high-performance devices to be designed, the thickness of magnet neutron shielding, particularly in the inboard leg, drops along with the size of the device. This is an inescapable issue since neutrons are the primary energy source in D-T fusion, but of course their trajectories are unaffected by the higher magnetic field. As neutron attenuation is largely an exponential, as opposed to linear effect, linear reductions in shield thickness lead to exponentially higher neutron fluxes reaching the magnet. In addition, the possibility of designing non-insulated HTS fusion magnets [30] makes the superconductor the primary radiation-limited material in such a magnet. Finally, in order to attain the high performance required for high-field magnets, the HTS in a compact device will be “sub-cooled” to temperatures between 10-20K, far below their critical temperatures [21]. These new conditions motivate an expansion of REBCO irradiation research to higher fluences, with measurements performed at the low temperatures and high fields which will be present in a compact, high-field design.

In order to assess the radiation survivability of the REBCO tapes in ARC, a model axisymmetric reactor geometry was studied using the Monte Carlo neutron transport code MCNP (see Figures 1-5 and 1-6). Using this model and assuming the ARC fusion power of 525 MW, the TF coils were found to reach the aforementioned $3 \times 10^{18}$ neutrons/cm$^2$ “limiting fluence” in just 10 full-power-years (FPY) of reactor operation. Although some 14.1 MeV neutrons will reach the magnet, a critical difference between the magnets and the first wall for radiation damage is that the majority of the high energy neutrons have been moderated by the time they reach the magnet. In fact, the normalized flux density vs. energy of the neutrons at the fusion reactor magnet in ARC look remarkably similar to a fission reactor core spectra (Figure 1-7). This is advantageous because it allows the use of research fission reactor facilities to simulate neutron damage to REBCO in fusion magnets, a technique employed in several studies [5, 6, 31]. Another advantage of fission reactor core irradiations is that the instantaneous neutron flux in a fission core is much higher than the flux reaching a fusion magnet, allowing fusion magnet lifetime fluences to be reached within days to weeks in a fission core.

The most recent radiation damage study on YBCO closest to ARC-relevant conditions has been performed to a maximum field of 15 T and maximum fluence of $3 \times 10^{18}$ n/cm$^2$ [6]. This study investigated changes of critical current density (the maximum current density allowed in a superconductor before it transitions to the normal state) with radiation. The critical current density is often used as a performance metric for superconductors in the context of building high-field magnets, since a higher current density allows more of the magnet volume to be taken up by structure, as opposed
Figure 1-5: Cross section of the axisymmetric geometry used to model neutron transport in ARC using MCNP. Yellow indicates the plasma region, orange indicates the TiH₂ shielding, light blue indicates FLiBe, and brown indicates the superconducting coil structure. Gray indicates either Inconel 718 or tungsten for the vacuum vessel, connection post, and divertor. The “squaring” of some corners in this simplified model has a negligible effect on the neutronics analysis.

to superconductor, thus allowing higher field magnets to be constructed. The study referenced above shows an initial rise of critical current with irradiation, followed by a drop at higher fluences, due to the competing influences of deleterious tape amorphization and the creation of favorable pinning sites as the REBCO is irradiated. This behavior has been observed in several previous studies as well [32, 33, 34]. The inflection point of the critical current trend occurs at higher fluences as the operating temperature of the REBCO is lowered, meaning that lower temperature REBCO operation improves “resistance” to radiation. At the study’s lowest measured temperature of 30 K (10 K higher than the operating temperature of the ARC magnets) and applied field of 15 T, the critical current at maximum fluence was still slightly higher than the unirradiated critical current.

Based on the above data, the ARC magnets were conservatively designed to a maximum lifetime fluence of $3 \times 10^{18} \text{ n/cm}^2$, ensuring that radiation damage will not be a concern to the REBCO in the device.

Given the space constraints of ARC, the molten salt FLiBe blanket alone was found to be insufficient as a neutron shield for the REBCO superconducting coils, particularly on the inboard side. Therefore, a shield composed of TiH₂ was around the
blanket tank (the component that holds the high-temperature molten FLiBe), with additional thickness on the inboard side. TiH$_2$ has an extremely high hydrogen density, appropriate for neutron moderation, and a high cross section for the neutron absorption, making it an ideal shielding material [35].

The ARC study concluded that after adding the TiH$_2$ shield, it would take 9 Full-Power Years (FPY) of reactor operation to reach a fluence of $3 \times 10^{18}$ neutrons/cm$^2$ at the limiting inner midplane region of the magnet. While this lifetime is likely insufficient for a dedicated, commercial-scale power plant, 9 FPY represents a conservative lower bound due to insufficient data on tape irradiation.

### 1.2.2 Motivation of research

While previous REBCO irradiation studies can be used to guide compact reactor design, the wide variety of tape production methods motivates large-scale irradiation studies to compare radiation effects on different tape compositions under compact reactor-relevant operating conditions such as cryogenic irradiation temperature. While such studies were performed on Nb$_3$Sn [36], no facilities currently exist which
Figure 1-7: Comparison of ARC neutron energy spectrum at the magnet coils with the in-core MIT Reactor fission neutron spectrum. The flux present at lower neutron energies for the ARC spectrum is due to the down-scattering of neutrons by the blanket and shield before reaching the magnet coil.

can perform cryogenic neutron irradiations at this time. Before such facilities were phased out in the 1990’s, a small number of cryogenic YBCO neutron irradiations were performed [37, 38]. While these studies showed differences between cryogenic and room temperature irradiations, there is no data at higher fluences and low temperature/high-field measurement conditions relevant to compact HTS fusion reactors. In addition, the irradiated YBCO samples were either single-crystal or sintered polycrystalline samples from the early days of HTS development as opposed to the long-length, high quality grain-aligned coated conductor superconductors available today.

As building new cryogenic neutron irradiation facilities will be an expensive undertaking, the purpose of this thesis is to determine whether radiation temperature is important from the point of view of future compact fusion-focused REBCO irradiation studies. For this work, protons were used to simulate the effect of neutron damage. Ion irradiation can be used as an effective screening tool, allowing large-scale scans to be performed on a wide range of tape compositions under a large number of irradiation conditions. At moderate energies, ion irradiation does not cause sample activation, significantly reducing the difficulty of analyzing the irradiated samples. Ion irradiation facilities at the required energies are considerably smaller, cheaper, and more flexible than research fission reactors, making specialized target chambers feasible in small, university labs. In addition, with moderate-current beams, ion irradiations can produce similar amounts of displacements per atom (DPA) to fission reactor irradiations in a fraction of the time.
The idea of using charged particle beams to emulate neutron damage has been investigated in the context of fission reactor material. Due to the high rates of DPA which can be obtained in self-ion irradiation, researchers have proposed using charged particle beams to test fission reactor structural material up to very high DPA, studies which would otherwise take tens of years in fission reactors [39]. The situation with HTS is somewhat different than the irradiation of fission structural materials in that only milli-DPA are required to begin observing macroscopic degradation (or enhancement) of tape performance. However, the fast pace of REBCO development over the past ten years, combined with the aforementioned plethora of tape production techniques and tape operating conditions makes the parameter space to be explored very large. Using ion beam irradiation as a screening tool will allow hundreds of permutations of tape composition and irradiation conditions to be explored, leaving a few promising candidates to be tested in much more resource and time-intensive fission reactor (and possibly fusion materials test facility) neutron irradiations.

In this work, several REBCO samples were irradiated at combinations of four different proton fluences and three different irradiation temperatures. These samples were then analyzed in a high-fidelity transport current measurement system to determine the effect that radiation temperature plays on $J_c$ degradation at high-fluences as well as the microstructural mechanisms behind the degradation at different conditions.

### 1.3 Goals and structure of thesis

This thesis has two main goals. The first goal of the thesis is to determine if irradiation temperature is important for a given fluence of proton irradiation. The second goal of this thesis is to use a combination of theory and simulation to determine whether the results of the cryogenic irradiations are applicable to neutron irradiations. The thesis is organized as follows: Chapter 1 introduces the motivations behind the research and outlines why this research is important. Chapter 2 outlines the basic theory behind superconductors and the mechanisms responsible for enhancement or degradation of critical current density with radiation. Chapter 3 describes the experimental methods for the REBCO irradiations. Chapter 4 describes the critical current characterization and data collection procedures. Chapter 5 presents an analysis of the critical current characterization measurements. Chapter 6 describes the Monte Carlo (MC) and molecular dynamics (MD) simulations performed to investigate the applicability of the Chapter 5 results to neutron irradiations. Finally, Chapter 7 provides concluding remarks about the applicability of the results to future design studies as well as suggestions for further work.
Chapter 2

Background

2.1 General superconducting history and theory

The first superconducting substance was discovered in 1911 by Heike Kamerlingh Onnes. While measuring the resistivity of ultra cold substances, Onnes noticed that the resistance of mercury dropped abruptly to zero at temperatures of less than 4.2 K [40]. This discovery marked the beginning of a rush to find more superconducting substances, and many elements were found to have a “critical temperature” at which they ceased to have a resistance. In the acceptance lecture for his 1913 Nobel Prize awarded for the discovery of superconductivity, Onnes discussed the possibility of using superconducting substances to "make feasible the production of strong magnetic fields using coils without iron" [41]. However, in the same lecture Onnes went on to describe two unexplained phenomena that might limit these possibilities – the loss of the superconducting state after threshold values of magnetic field and current density were reached in the superconducting material. The values of these “critical fields” and “critical current densities” would become the two other defining parameters (along with the aforementioned critical temperature) defining whether a substance was superconducting or not. These three critical parameters are dependent upon each other and can be represented for each superconducting material as a “critical surface” in three dimensions, with the axes of temperature, current density, and magnetic field (see Figure 2-1).

It was quickly discovered that all purely elemental superconductors would enter the normal state at very low current densities and magnetic fields, rendering pure metals unsatisfactory for the construction of practical magnets. However, after all practical pure elements had been tried, scientists began experimenting with alloys. In 1933, Meissner and Ochsenfeld discovered that once a substance in a background magnetic field became superconducting, it would not perfectly freeze in the magnetic field (as would be expected of a purely conducting material) but rather would expel the magnetic field by creating a screening current on its surface to become a perfect
Figure 2-1: Schematic of a critical surface of a superconductor [42]. The shape of the surface is caused by the dependences of $J_c$, $B_c$ and $T_c$ on each other and is different for each type of superconductor.

diamagnet [43]. This effect, which would become known as the Meissner Effect, can be observed in all superconductors. Shortly thereafter, the London brothers postulated that a pure surface current would imply infinite current density, which would be impossible. Instead, they proposed finite field penetration into the superconductor’s surface, allowing for realistic screening current densities [44]. This field penetration was observed to have an exponential character, with characteristic e-folding length ($\lambda$) known as the penetration depth, defined as:

$$B(x) = B_0 \exp \left( \frac{-x}{\lambda} \right)$$

(2.1)

where $B_0$ is the applied field and $x$ is the depth into the superconductor. Typical values of $\lambda$ are on the order of 10-100 nm for pure metals and several hundred nm for high-temperature superconductors [42].

Shortly after the discovery of the Meissner effect, Gorter and Casimir put forth a first-principles theory on the microscopic mechanisms behind the superconducting state. In this theory, they postulated a two-fluid model of normal electrons and superelectrons [45]. The two-fluid theory was useful for the development of the idea of an order parameter relating the superelectrons proposed by Ginzburg and Landau in 1950 [46]. In 1953, Pippard further solidified the idea of the order parameter. Based on the earlier work of Gorter and Casimir, Pippard hypothesized that superconductivity was a quantum mechanical "coherent effect" between the superelectrons [47]. Pippard defined a parameter called the “coherence length” ($\xi$), the characteristic
Table 2.1: Coherence lengths in YBCO for different temperatures and orientations. The subscripts indicate the directionality of the coherence length with respect to the crystallographic axes of the superconductor.

<table>
<thead>
<tr>
<th>Temperature [K]</th>
<th>$\xi_{ab}$ [nm]</th>
<th>$\xi_c$ [nm]</th>
</tr>
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<tbody>
<tr>
<td>0</td>
<td>1.7</td>
<td>0.3</td>
</tr>
<tr>
<td>77</td>
<td>3.5</td>
<td>0.7</td>
</tr>
</tbody>
</table>

distance over which a majority of the electrons in a material become superelectrons, essentially defining a length scale for the boundary between normal and superconducting phases. Pippard observed that the coherence length was dependent on the direction of the current in the material, leading to different coherence lengths for the different orientations of crystal axes in anisotropic materials. In addition, Pippard observed that the coherence length decreased as impurities were added to a pure superconducting metal, due to the decrease of the electron mean free path ($l_e$) as impurities were added, so that coherence length could be expressed as [48]:

$$\xi \approx \sqrt{\xi_0 l_e}$$  \hspace{1cm} (2.2)

Both $\xi$ and $\lambda$ have the following dependencies on $T$:

$$\xi = \xi_0 \sqrt{\frac{1}{1 - \frac{T}{T_c}}} \quad \lambda = \lambda_0 \sqrt{\frac{1}{1 - \frac{T}{T_c}}}$$  \hspace{1cm} (2.3)

The work of Ginzburg, Landau, and Pippard was followed shortly by L.N. Cooper’s theory that below a certain temperature, the superelectrons predicted by Pippard would become paired due to the electron-phonon effect [49]. These electron pairs (referred to later as “Cooper pairs”) had a collective integer spin, thus behaving as bosons instead of fermions and able to all occupy the same energy state and acted along the length scale of $\xi$. The idea of Cooper pairs was formalized into the Bardeen, Cooper, and Schrieffer (BCS) theory, the first microscopic theory to describe metallic superconductors [50]. While high-temperature superconductors have not been fully explained by any theory to date, theories have been developed to approximate the behavior of HTS [51, 52] and many of the concepts from BCS theory still apply (such as the coherence length and penetration depth). Approximate values of $\xi$ for YBCO are shown in Table 2.1 [42].

Thermodynamically, the penetration depth and coherence length are both related to the free energy density of the superconductor. In the presence of an applied magnetic field, there is an increase in free energy density due to magnetization which takes place along a distance on the order of the penetration depth. However, there is also a decrease in free energy density from the electron ordering which takes place along a distance on the order of the coherence length. The free energy density magnitudes
of both contributions far into the superconductor are given by [53]:

$$|\Delta g| = \frac{B^2_c}{2\mu_0}$$  \hspace{1cm} (2.4)

For distances $> \lambda, \xi$, the contributions from both effects identically cancel out. However, close to the superconducting/normal boundary, the change in free energy from both effects is unequal. When $\lambda$ and $\xi$ are unequal, this leads to a net positive or negative surface energy close to the boundary depending on whether $\lambda$ is larger (negative surface energy) or $\xi$ is larger (positive surface energy). The energy density difference as a function of length near the surface can be approximately given as:

$$\Delta g(x) \approx \frac{B^2_c x}{2\mu_0(\lambda - \xi)}$$  \hspace{1cm} (2.5)

where $B_c$ is the critical field. The sign of this energy difference is important to determining the stability of the superconducting phase (see Fig. 2-2). If the surface energy near the normal-superconducting boundary is positive, the superconducting state is thermodynamically stable throughout the whole material until the critical field is exceeded. This class of superconductors is referred to as “Type I” superconductors. However, if the surface energy near the normal-superconducting boundary is negative, this means that it is thermodynamically favorable for part of the superconductor to transition to the normal state.

This sets up a situation where volumes of normal material interspersed throughout the superconductor appear and allow magnetic flux lines to pass through them. This second class of superconductors is referred to as “Type II” superconductors and is characterized by having an upper and lower critical field, as opposed to a single critical field like Type I superconductors. Below the lower critical field, Type II superconductors behave as Type I superconductors and expel all magnetic flux. However, past the lower critical field but below the upper critical field, Type II superconductors allow magnetic flux (quantized in units of $\Phi_0 = h/2e \approx 2 \times 10^{-15}$ T m$^2$) to penetrate normal volumes in the superconductor in a “mixed state” (also called the Shubnikov state). In order to shield the rest of the superconductor from the penetrating fields, screening vortex currents flow around the normal cores to oppose the applied field. Normal cores have a radius on the order of $\xi$ and the screening currents flow around the normal cores at a radius on the order of $\lambda$. It is thermodynamically favorable for these normal cores to arrange themselves in a triangular lattice throughout the Type II superconductor (depicted in Figure 2-3). For HTS with an anisotropic crystal structure (such as REBCO), the different values of $\xi$ and $\lambda$ depending on the direction of the crystallographic axis lead to normal cores of different shapes, as shown in Figure 2-4.

As the applied magnetic field increases, the lattice of normal cores grows denser until
Figure 2-2: Schematics of free energy density contributions from electron ordering (negative contribution with a length scale of $\xi$) and magnetization (positive contribution with a length scale of $\lambda$) \cite{48}. The sum of the two contributions determines whether the boundary between superconducting and normal phases is thermodynamically stable, expelling all magnetic flux (Type I superconductor) or unstable, allowing some flux to penetrate (Type II superconductor).

When the cores begin to overlap and the bulk superconducting state is lost throughout the material. The point at which this occurs is the aforementioned upper critical field. It is important to note that because applied magnetic flux is able to pass through the superconductor, the upper critical field of a Type II superconductor is much higher than the critical field of a Type I superconductor, thus making the construction of electromagnets with Type II superconductors feasible. If both $\lambda$ and $\xi$ are known, the Ginzburg-Landau parameter $\kappa = \lambda / \xi$ can be used to classify whether a superconductor is Type I or Type II. If $\kappa \geq 0.71$ the superconductor will be of Type II and if $\kappa \leq 0.71$, the superconductor will be Type I \cite{56}.

The property of Type II superconductors of being able to retain their superconducting state at much higher fields has the effect of allowing them to carry more current. The magnetostatic version of Ampere’s Law applied to current flowing through a wire of cross sectional area $A$ is:

$$B = \frac{\mu_0 J A}{2 \pi r}$$  \hspace{1cm} (2.6)
Figure 2-3: SQUID microscopy image of flux vortices in YBCO [54]. It is important to note that lack of resolution on the SQUID microscope is responsible for the larger-than-expected vortex diameters in this image. Nevertheless, the centers of the vortices should be accurate and thus the distribution (i.e. approximately triangular lattice) of the flux vortices is experimentally observed.

Figure 2-4: Schematic of normal core and vortex dimensions in an anisotropic superconductor such as REBCO where $\xi$ and $\lambda$ have different values depending on which crystallographic axis is being investigated [55]. Normal cores are characterized by $\xi$ and vortex current loops are characterized by $\lambda$. 
where \( J \) is the current density flowing axially down a wire and \( B \) is the magnetic field at radius \( r \) from the center of the wire. Thus, even if no external field is applied to a superconductor, it will necessarily feel a “self-field” if current is passed through it. For a Type I superconductor, the critical current was found to be the value at which the self-field produced by the applied current matched the critical field \([57]\). Since a Type II superconductor has a much higher critical field (due to its property of allowing some magnetic flux to penetrate the material), it therefore also has a much higher critical current at a given temperature.

### 2.2 High temperature superconductors

While certain low-temperature superconductors such as NbTi and Nb\(_3\)Sn are Type II superconductors, this thesis will focus on high-temperature cuprate superconductors, particularly REBCO. In 1986, J. Georg Bednorz and K. Alex Muller \([58]\) discovered a superconducting material with a critical temperature near 35 K, far higher than any previous critical temperature observed. This material, a lanthanum cuprate with barium doping, was a ceramic, anisotropic perovskite structure. Shortly thereafter another cuprate with a critical temperature of 93 K was discovered experimentally by Paul Chu \([59]\). This compound, called YBCO (an acronym for, yttrium, barium, copper, and oxygen) has been aggressively investigated in the past 30 years and expanded into the more general category of REBCO, with other rare earth metals (RE) being substituted for yttrium. While much of the detailed, early work on high-temperature cuprates was performed on YBCO, the results of these studies are generally applicable to all RE cuprates, and the more general term “REBCO” is used throughout this thesis.

The chemical structure of REBCO is shown in Figure 2-5, with a basic unit cell on the left and the more commonly depicted repeat structure showing the “stacked planes” of the superconductor on the right. The CuO\(_2\) plane is responsible for carrying the supercurrent, and the Cu-O chain layers act as a charge reservoir for the the CuO\(_2\) plane. Both of these planes will become important later when discussing pinning.

In addition to having a much higher critical temperature (leading to the classification of “high temperature superconductor”) than traditional superconductors, high temperature superconductors were found to have a much higher critical field and critical current (Figure 2-6). Early excitement over the technological possibilities afforded by high temperature superconductors was quickly dashed when it was discovered that grain boundary orientation between YBCO single crystals was important to preserving the high critical current densities achievable in the single crystals. The techniques to fabricate large samples of high-quality YBCO into wires would take 10 years to develop and another 10 years to commercialize. This large dependence of critical current on grain boundaries was later found to play a role in the effect of radiation damage in superconductors, as will be shown below.
Figure 2-5: Orthorhombic structure of the REBCO unit cell, generated with the VESTA 3D visualization program [60] on the left, along with a repeated structure to illustrate the three different superconducting planes (each of which repeats once in the c-direction per unit cell) on the right. Oxygen is red, copper is blue, barium is green, and the rare earth element is gray. The highly anisotropic structure of REBCO contributes to the differences in critical current at different applied field angles.

Figure 2-6: Comparison of LTS (red) and HTS (green) critical surfaces, demonstrating the order-of-magnitude increase in all three parameters in HTS [courtesy of D. Brunner].
Figure 2-7: Schematic view of the cross section of REBCO tape superconductor by SuperPower Inc. Each layer is shown with its typical thickness. The tape is normally available in widths between 2 and 12 mm, and a total thickness of between 0.05 to 0.1 mm [courtesy of F. Mangiarotti].

Fig. 2-7 shows a schematic view of the cross section of commercially available REBCO tape conductor by SuperPower Inc. The conductor is mostly composed of copper and Hastelloy, with a very thin layer (approximately 1% of the total thickness) of REBCO superconductor [61]. The buffer layers are used during manufacturing to orient the REBCO crystals in a preferred direction, such that the c-axis of the crystals are perpendicular to the face of the tape [62] (note that the orientations of Figure 2-5 and 2-7 are the same). The critical current density of REBCO is very sensitive to the orientation of the magnetic field due to the anisotropy of the superconducting lattice.

### 2.2.1 Flux pinning

The final way that irradiation can influence superconductor properties is through flux pinning effects. The property of Type II superconductors allowing lines of magnetic flux to penetrate the superconductor has important implications to the critical current. As current is passed through the superconductor, the flux lines in the normal cores will experience a Lorentz force:

\[
\vec{F}_L = \vec{J} \times \vec{B}
\]

This force will cause the flux lines to move and will only be counteracted by a “pinning” force exerted by defects in the superconducting material which hold the flux lines in place as shown in Figure 2-8. The mechanism responsible for the pinning force is the same thermodynamic mechanism responsible for flux inclusion of Type II superconductors described above. In a Type II superconductor, it is energetically favorable for flux lines from an applied magnetic field (above the first critical field) to penetrate some of the superconducting regions, transforming the pure superconductor into a mixed state of superconducting and normal regions. In this way, the system reaches equilibrium when a certain amount of flux has penetrated the superconduc-
Figure 2-8: Schematic of normal core formation and pinning site force contributions which counteract the Lorentz force on the flux lines when current is passed through the superconductor.

The first pinning classification is simply whether the pinning sites are intrinsic to the ideal superconducting lattice or whether they are extrinsic, or artificially produced. In REBCO, the large asymmetry between current-carrying capacity depending on field orientation arises from the fact that the Cu-O chain layers act as 2D intrinsic pinning centers to magnetic flux lines parallel to the ab plane of the tape [63]. For this work, the focus will be on artificial pinning centers, such as point defects or extended defects introduced by irradiating the superconductor, although it will be seen that the interaction of radiation with intrinsic pinning centers causes changes in the angular dependence of irradiated $J_c$ measurements.

The second pinning classification deals with the length over which the pinning force operates. The two most important length scales relating to flux vortices are the aforementioned penetration depth ($\lambda$) and coherence length ($\xi$) where $\lambda$ is the length scale for the screening current vortices and $\xi$ is the length scale for the normal cores [56]. For YBCO, since $\lambda$ is approximately 100 times the value of $\xi$, it is more effective to introduce pins on the order of $\xi$ ($\sim$ 1-4 nm) to maximize the pinning density, so manufacturers often introduce artificial sites (such as BZO nanorods or RE$_2$O$_3$ precipitates) on the order of a few nm to increase performance. In the context of radiation damage, many of the observed defect clusters [5] created in REBCO by neutron irradiation are on the order of several nanometers, comparable to $\xi$.

The third classification is the strength of the pins. Strong pinning sites distort the flux line lattice itself, and are generally very stable against thermal motion from the vibration of lattice atoms even at high temperatures (see Figure 2-9 a). Weak pinning sites act collectively, and the shape of the flux line lattice is preserved (see Figure 2-9 b). An example of a strong pinning site would be a BZO nanorod or...
Weak pinning sites are smaller in size, such as point defects or small defect clusters. The volumetric pinning force associated with strong pins can be expressed as [64]:

\[ F_{p,s} = n_{a,f} f_{p,l} \]  \hspace{1cm} (2.8)

where \( n_{a,f} \) represents the areal flux vortex density and \( f_{p,l} \) is the pinning force per unit length along the pinned flux line. The weak pinning force density can be expressed as:

\[ F_{p,w} = \left( \frac{n_{v,w} f_{p,0}^2}{V_c} \right)^{1/2} \]  \hspace{1cm} (2.9)

where \( n_{v,w} \) is the volume density of weak pins, \( f_{p,0} \) is the force per weak pin, and \( V_c \) is the volume of weak pins acting to pin a single flux line. It is important to note that the nomenclature “strong” and “weak” refers to the strength of the pinning sites relative to the thermal fluctuations in the lattice, not necessarily to their effectiveness at pinning stronger or weaker applied fields. In fact, due to their collective nature, “weak” pinning sites are often dominant at high field conditions because thermal motion is suppressed at cryogenic temperatures.

Figure 2-9: Schematics of strong and weak pinning sites. The strong pinning site deforms the shape of the flux line, while the weak pinning sites act collectively to pin a flux line and it retains its shape.

The fourth classification is the directionality of the pinning site. Random pinning centers have no particular direction and increase \( J_c \) at all applied field angles. However, correlated pinning sites act primarily to pin flux lines in one orientation. For example, BZO nanorods introduced parallel to the c axis of the superconductor will effectively pin fields applied parallel to the c-axis but their pinning effectiveness vanishes at other angles (Figure 2-10). This effect is diminished at lower temperatures and high fields.
Figure 2-10: $J_c$ vs. $\theta$ plot showing correlated pinning at 0, 90, and 180 degrees to the $ab$ plane. The 90 degree peak arises due to the intrinsic Cu-O chain layer pinning and the 0 and 180 degree peak arises due to the 1D BZO nanorod artificial pinning sites oriented along the c-axis of the superconductor.

but underscores the importance of obtaining angularly-resolved $J_c$ measurements for the purposes of designing fusion magnets where twisted cable configurations typically used to prevent AC losses and the minimum $J_c$ is limiting [65].

The final pin classification is the dimensionality of the pinning site. Point defects are considered 0D pinning sites, and all larger pinning sites can be classified as being 1D, 2D, or 3D. Examples of the larger pinning sites would be BZO columns as 1D defects, Cu-O chain layers as 2D defects, and RE$_2$O$_3$ precipitates as 3D defects, as shown in Figure 2-11.

Radiation of superconductors can lead to both enhancement and degradation of $J_c$ through pinning effects (discussed in detail in Section 2.3). The creation of defects due to irradiation introduces new pinning sites to the superconductor, leading to enhanced transport current in the pinning-dominated regime. Neutron irradiation has been shown to create amorphous defect clusters on the order of several nanometers (and thus, the coherence length) [5], while light ion and electron irradiation produce mainly point defects [66]. The operational regime (i.e. operating temperature, field, and field angle) at which pinning effects are beneficial depends on the character of the introduced defects related to the pinning characteristics discussed above. In addition to the enhancement effect of the creation of new pinning sites themselves, there is some evidence that irradiation-introduced pinning centers can synergistically interact with already-existing pinning sites to produce added $J_c$ enhancement [67].

The deleterious effect of irradiation on pinning is that oxygen displacements will cause disorder in the CuO$_2$ plane, which distorts the correlated pinning effects from the Cu-
Figure 2-11: TEM images showing examples of 1D, 2D, and 3D pinning sites (0D sites are too small to be resolved with a TEM): 2-11a BZO nanorods, 2-11b Cu-O chain layers, and 2-11c 3D precipitates. Examples of pinning sites are pointed out or shaded in blue to guide the eye.

O chain layers at field angles close to 90 degrees from the \( ab \) plane\(^1\). It has been well established [63] that the anisotropic nature of the critical current in REBCO is due to the intrinsic pinning effect of the Cu-O chains, which have spacing on the order of \( \xi \). The non-superconducting Cu-O chain layers allow flux lines to penetrate, but these flux lines are sandwiched in between the superconducting CuO\(_2\) planes, and effectively pinned [5]. As the CuO\(_2\) plane are disordered, normal regions will develop in the planar surfaces which allow flux lines to hop between planes, reducing the intrinsic pinning effect, and thus \( J_c \) for fields applied parallel to the \( ab \) plane.

\(^1\)Due to varying notation in the literature, it is important to define the angular orientations discussed in this paper. Angles of 0 and 180 degrees refer to a field angle parallel to the \( c \)-axis of the superconducting lattice (i.e. perpendicular to the flat part of a macroscopic coated conductor tape) whereas angles of 90 and 270 degrees refer to a field angle parallel to the \( ab \) plane of the superconducting lattice.


2.2.2 Matching field

Since magnetic flux penetrating a Type II superconductor is quantized into bundles of $\Phi_0 = h/2e \approx 2 \times 10^{-15}$ T m$^2$, the areal density of these flux lines (also known as flux vortices) can be expressed as:

$$n_{a,f} = \frac{B}{\Phi_0} \quad (2.10)$$

where $B$ is the applied field on the superconductor. As seen in Figure 2-3, the flux vortices will organize themselves into the most efficient areal packing scheme, which is a triangular lattice. The packing efficiency of a triangular lattice is $\eta_{2D} \approx 0.91$, so the density of flux lines can also be expressed as:

$$n_{a,f} = \frac{\eta_{2D}}{\frac{\pi}{4} a^2} \quad (2.11)$$

where $a$ is the mean spacing between flux lines. Combining Eq. 2.10 and 2.12, the field-dependent mean spacing can be expressed as:

$$a = \sqrt{\frac{\eta_{2D} \Phi_0}{\frac{\pi}{4} B}} \quad (2.12)$$

In order to obtain optimum pinning at a given field, an ideal superconductor would have one pinning site per flux vortex. Thus, the concept of a “matching field” ($B_m$) can be established. If $n_{a,p}$ represents the areal density of effective pinning sites, the matching field can be expressed as:

$$B_m = n_{a,d} \Phi_0 \quad (2.13)$$

It is important to emphasize that $n_{a,d}$ represents the areal density of effective pinning sites, and is thus a complicated function of operating field, operating field angle, operating temperature, tape preparation, and irradiation conditions.

2.3 Radiation effects in superconductors

In the context of superconductor performance for fusion magnets, the critical current, $I_c$, is the figure of merit for assessing “damage” or “enhancement” to the superconductor. Although $I_c$ is the directly measurable quantity, the microscopic physics performance parameter is really the critical current density through the cross section.
of the superconductor itself (neglecting the addition of mechanical and stabilizing layers present in commercial tapes). This quantity, $J_c$, can be determined by dividing the measured $I_c$ by the width of the coated conductor and the thickness of the REBCO layer (determined by TEM measurements), and will be used as the tape performance metric throughout the rest of this paper.

As REBCO is irradiated, a wide variety of changes occur within the ordered superconducting crystal lattice and competing effects on $J_c$ emerge. On one hand, the displacements of atoms and creation of defect clusters has the tendency to lower $J_c$ through $T_c$ suppression, lattice amorphization, degradation of intergrain current transfer, and the disordering of intrinsic pinning sites [68]. On the other hand, point defects and defect clusters can act as artificial pinning centers, increasing $J_c$ by the creation of beneficial pinning sites [69]. The cumulative effect of these mechanisms can be observed as a net increase or decrease of measured $J_c$. A summary of these mechanisms is presented here.

### 2.3.1 $T_c$ suppression with radiation

The first mechanism of $J_c$ degradation in irradiated superconductors comes through suppression of $T_c$ by the means of the accumulation of radiation-induced defects. Near $T_c$, the dependence of $J_c$ on temperature can be given by Ginzburg-Landau (GL) theory as [70]:

$$J_c = J_0 \left(1 - \frac{T}{T_c}\right)^{3/2}$$  \hspace{1cm} (2.14)

Thus, a degradation of $T_c$ will lead to a reduction of critical current. The chemical reason for $T_c$ degradation was discovered to be related to the oxygen deficiency $\delta$ of $\text{Y}_1\text{B}_2\text{Cu}_3\text{O}_{7-\delta}$ [71], which was later discovered to be a measure of the “doped hole concentration” $p$, defined as the concentration of holes per Cu atom in the CuO$_2$ planes [72]. It was found that $T_c$ varied parabolically with hole concentration, with an optimum hole concentration of $p \approx 0.16$ as seen in Figure 2-12.

The explanation for the effect of hole concentration on $T_c$ has been theorized to be enhanced electron scattering due to the added magnetic and non-magnetic impurities leading to “de-pairing” of the Cooper pairs carrying the supercurrent. This scattering occurs in the superconducting CuO$_2$ planes as more Cu and O vacancies are introduced. In a systematic study of defects introduced through electron irradiation [73], it was found that Cu vacancies are approximately twice as effective at O vacancies at pair breaking. This study also determined that irradiation-induced $T_c$ degradation for the electron irradiations was insensitive to radiation temperature below 300 K, although it should be noted that the study did not investigate the ef-
fects of higher-temperature irradiation up to $\sim 400$ K which has been the case in more recent accelerator and fission reactor irradiations.

The other microstructural explanation for pair breaking is related to the structure of the entire cuprate lattice. As mentioned above, the oxygen stoichiometry of cuprate superconductors was found to have a large influence on the lattice parameters and structure of the superconducting crystal [71]. At a $\delta$ value of approximately 0.6, a phase transition from tetragonal to orthorhombic occurs in the crystal lattice of the cuprate [74], with the tetragonal structure being non-superconducting and the orthorhombic structure being superconducting. This led to the development of a “charge reservoir/transfer” model where Cu-O chains act as reservoirs for charge at the CuO$_2$ superconducting planes [75]. The disorder of the Cu-O chains affects the number of electron holes (i.e. charge carriers) available to the CuO$_2$ planes [76]. It is important to note that radiation damage leading to $T_c$ degradation is distinctly different from full amorphization of the superconducting lattice. The damage to the CuO$_2$ planes which causes $T_c$ does not fully amorphize the region, and reduced supercurrent can still flow along the planes. From a microstructural point of view, the type of damage leading to $T_c$ degradation would be point defects or small clusters of point defects.

### 2.3.2 Lattice disorder

The second general type of radiation damage that can occur in superconductors is full amorphization of the superconducting lattice, leading to normal regions within the superconductor. The filamentary model of Moeckly et al. [77] describes high temperature superconductors as, “a composite system consisting of a network of superconductive filaments embedded in a nonsuperconductive matrix”, with the size of the filaments being on the order of the superconducting coherence length, $\xi$. As defect
clusters and cascades are introduced into the superconducting material, the network of superconducting filaments becomes less and less dense until the superconducting state collapses completely.

Early experiments by Kirk and Frischerz [78, 79] observed (through TEM measurements) the formation of an intragranular “cellular” microstructure of superconducting cells with a diameter of ~5-10 nm surrounded by highly amorphous regions in irradiated YBCO crystals. The onset of this microstructure corresponded to a rapid degradation of $T_c$ (and thus $J_c$) suggesting the complete blockage of supercurrent by these cellular boundaries. An interesting observation of these studies was that the onset of the cellular structure was highly dependent on both the original superconductor quality (defined as the sharpness of transition between superconducting and normal state, characterized by the “n-value” as described in Chapter 4) and the type of irradiation. Cellular onset was observed at a calculated DPA of 0.07 for ion irradiations but only 0.003 for neutron irradiations, suggesting that this structure was only created by the large cascades produced by the high-energy recoils characteristic of neutron irradiation [38]. Later experiments with higher quality YBCO crystals did not observe the onset of cellular microstructure for fluences up to $8 \times 10^{17}$ n/cm$^2$ and hypothesized that it would require fluences on the order of $\approx 5 - 10 \times 10^{18}$ n/cm$^2$ to observe the onset of the cellular microstructure, based on high-fluence ion irradiations of the improved YBCO crystals.

### 2.3.3 Grain boundary disorder

For modern coated conductors such as commercial REBCO tapes, it is important to make the distinction between critical current within grains and between grains. Grain boundary misorientations of even a few degrees lead to bulk degradation of the sample $J_c$ of factors of 10 to 50, and the application of magnetic fields only increase this deleterious effect [80, 81]. The microstructural mechanisms between lattice (intragrain) and grain boundary (intergrain) current degradation are similar and related to weak coupling between the superconducting filaments [82].

Weak coupling arises due to the “Josephson effect” [83], a macroscopic manifestation of the quantum mechanical nature of superconductors. The Josephson effect is the property of supercurrent to flow through a thin (~1 nm) insulating layer (referred to as a Josephson Junction) due to quantum tunneling and has been extensively studied since the 1960’s [42], particularly in the application to Superconducting Quantum Interference Devices (SQUIDs). Within the filamentary model of HTS superconductors, filaments can be connected by these weak links, allowing the so-called “glassy” superconducting phase to occur within a single grain [84]. Early experiments showed evidence for two distinct contributions to critical current density, a “weak link coupling”-dominated contribution and a pinning contribution [82], the former contribution being dominant at low fields and the latter contribution being dominant at high fields. While intragrain weak coupling was shown to exist at extremely low
fields, the more relevant weak coupling regime for fusion applications exists for intergrain (grain boundary) coupling, which can exist up to fields of several Tesla in unirradiated superconductors [85].

Typical grain boundaries in modern coated conductors are on the order of a nanometer (see Figure 2-13), similar to the superelectron coherence length for YBCO as shown in Table 2.1. As a bulk HTS coated conductor is irradiated, its grain boundaries will act as sinks to defects and widen, becoming progressively stronger barriers to transport current as the grain exceeds the coherence length. Current transport through a grain boundary can be calculated as [86]:

\[
J_c = J_0 \exp(-2\kappa \Delta)
\]  

(2.15)

where \(J_c\) is the tunneling current density through the boundary, \(J_0\) is the current density at the boundary, \(\kappa\) is the decay constant, given by [86] as 7.7 nm\(^{-1}\), and \(\Delta\) is the width of the boundary interface. Thus, as the HTS is irradiated, weak link coupling due to grain boundary widening will become the dominant effect limiting critical current density at higher and higher fields as \(J_c\) drops below the pinning-limited \(J_c\). Recent work on neutron-irradiated superconductors [31] has shown that the crossover in transport \(J_c\) vs. B curves for irradiated and unirradiated coated conductors can be used to approximately determine the field at which critical current changes from a grain-boundary-limited regime to a flux-pinning-limited regime.

Figure 2-13: TEM image of a 30 degree [001] tilt grain boundary in YBCO. The structural units of the grain boundary have been highlighted in color to show the approximate width (~1 nm) of the grain boundary [87].
2.4 Comparing ion and neutron damage mechanisms

A recent body of work indicates that under certain conditions, charged particle irradiation can be used to emulate neutron damage, and this technique is being explored as a possibility for many different types of nuclear materials testing [88, 89]. It has been proposed that varying the irradiation temperature of a material or irradiation flux could allow one to adjust the defect formation kinematics during ion irradiation in order to emulate the effects of neutron irradiation [39].

The use of accelerator-based, charged particle irradiation to emulate neutron damage of HTS tapes has several advantages. Accelerator systems in the desired ~MeV energy range are orders of magnitude smaller and cheaper than fission reactors, allowing research to be performed in small-scale, university settings such as MIT. Charged particle irradiations produce damage faster and without associated material activation of neutron irradiation. Finally, accelerator-based experiments allow for much more complicated irradiation setups, enabling the HTS samples to be irradiated while at cryogenic temperatures and coil-relevant strains. This combination of fast experimental turn-around-time and flexibility will allow for a large and varied set of HTS damage experiments to be performed, enabling the determination of HTS lifetimes over a large parameter space.
Chapter 3

REBCO Superconductor Irradiation

To test for the effects of irradiation temperature on $J_c$ degradation, a series of REBCO samples were irradiated with protons using a temperature-controlled target stage. The stage could maintain constant target temperatures from 80 K to 423 K, allowing a range of irradiation temperatures to be investigated. The following chapter describes the operation of and repairs on the tandem electrostatic accelerator used to perform the proton irradiations as well as details of the target chamber and irradiation conditions.

3.1 The DANTE tandem accelerator

3.1.1 General description and operation of accelerator

In order to perform REBCO ion irradiations, the DANTE accelerator (see Figure 3-1) was used at the MIT CSTAR facility. DANTE is a high-current tandem electrostatic accelerator, designed by Newton Scientific Industries in Cambridge, MA. The accelerator’s multicusp source and extraction optics allow for high DC beam currents, providing up to tens of $\mu$A of beam current at the high-energy end of the accelerator. The acceleration terminal can be charged up to 0.6 MV, allowing for 1.2 MeV proton/deuterium beams. While a complete description of the accelerator can be found in the Master’s thesis work of B. Blackburn [90], the general description and principles of operation are summarized below.

Ion source

DANTE utilizes a multicusp “bucket” source to produce negative ions [91] (Figure 3-2). The source operation is as follows: A high current (150 A) is passed through a tungsten filament at the front of the source, causing the filament to heat to tem-
temperatures of approximately 2000 K (1727 °C) and thermionically emit electrons. The filament is biased at -110 V with respect to the inside of the source body, accelerating the electrons away from the filament into the first magnetic volume of the source. The ~100 eV electrons are confined by the multicusp field around the edges of the source body produced by a configuration of permanent magnets as shown in Figure 3-3. At the same time, a small amount of source gas is introduced into the first magnetic volume, which is ionized by the electrons. The source is separated into two magnetic volumes by reversing the polarity of two of the multicusp magnets halfway down the source, creating a straight field line section across the cross section of the source body, often referred to as the magnetic source filter [92]. This straight field confines the electrons along its length, essentially creating a transport barrier down the middle of the source. Since the transport across this barrier (i.e. axially down the source) is controlled by collisional transport, the diffusion of electrons is proportional to $1/T^2$ [1], effectively separating the two regions into a high-energy electron region (the first region with the filament) and a low-energy electron region (the second region after the filter). In the second region, a process called dissociative attachment [93] allows electrons to bond with vibrationally excited protons, creating negative ions. The magnetic volumes are separated so that the ionization cross section for nuclei is maximized in the first volume at ~100 eV, while the dissociative attachment cross section is maximized in the second volume at ~1 eV.

After the negative ions have been produced in the second magnetic volume, the plasma electrode, which is biased 10 V above the source body walls allows the negatively charged nuclei as well as electrons to drift out of the source body into the extraction volume. In the extraction volume, a shaped extraction electrode is biased 1 kV above the plasma electrode, accelerating the negative ions and electrons to 1 keV. A weak magnetic filter in the extraction electrode steers the electrons into the electrode
itself while the negative ions continue through the electrode due to their much lower relative velocity. The extracted ions then pass through an Einzel lens, which focuses the beam into a tight beamspot, as well as accelerating it to approximately 10 keV before entering the acceleration column.

**Acceleration column**

The main acceleration column is a 3 m long tube, whose central position is a high voltage (0.6 MV) terminal. In order to produce a uniform accelerating gradient, the terminal voltage is stepped down to ground via a series of 200 MΩ resistive voltage divider steps. There are 47 stages on the low energy side of the column and 52 stages at the high energy side of the column. As the negative ions move through the first half of the column, they are attracted to the higher positive potential and gain energy. By the time the ions reach the terminal, they have an energy equal to their original charge state multiplied by the terminal voltage. At the terminal, there is a very thin (50 nm) carbon foil which the beam of negative ions passes through. At beam energies above \( \sim 100 \text{ keV} \), the charge exchange cross section starts to drop below the ionization cross section and the foil effectively changes the negative ion beam into a positive ion beam. The beam ions (which are now positive) are repelled from the positive terminal potential down the second half of the acceleration column. The final energy of the beam can be represented as:

\[
E_{\text{beam}} = |q_i| V_t + |q_f| V_t
\]  

(3.1)

where \(|q_i|\) is the initial charge state, \(|q_f|\) is the final charge state, and \(V_t\) is the terminal voltage. For hydrogen and deuterium beams, \(|q_i| = |q_f| = 1\), so the final beam energy is equal to \(2V_t\).
Focusing and steering

After exiting the acceleration column, the high-energy beam passes through a set of magnetic focusing quadrupoles and a set of steering magnets. Magnetic quadrupoles are used instead of an electric Einzel lens system (as in the source) because the high velocity of the beam particles would make the required electric field to deflect the particle trajectories impractical, whereas the magnetic force deflecting the particles is linearly dependent on the velocity of the particles:

\[ F_L = q\vec{E} + q\vec{v} \times \vec{B} \]  

The steering magnets simply create a straight field in either the horizontal or vertical direction to allow movement of the beam in the X/Y plane. After the beam passes through the steering and focusing magnets, it goes through a large field selection magnet which allows the beam to be steered horizontally to one of four beamlines at different angles from the straight beam path. In addition to allowing multiple beamlines to be set up at the same time, the selection magnet ensures that only particles with the same charge/mass ratio are steered into the desired beamline, eliminating “stray” beam of other atomic species and/or energies arising from unavoidable contaminant gases in the source.
3.1.2 Accelerator Repairs and Refurbishment

Significant refurbishment was required before using the accelerator to irradiate HTS. The two main systems needing refurbishment were the high-voltage system for the acceleration column and the ion source.

High Voltage System

The high terminal potentials required for acceleration require voltages in the range of 100's of kV, which presents an arcing hazard. In order to mitigate this hazard, the acceleration column is surrounded by a high-strength aluminum tank pressurized to ~70 psig with sulfur hexafluoride (SF₆), a dielectric gas. Nevertheless, arcs still can occur if the terminal voltage is raised too rapidly or too high. Past arcs had damaged the high voltage system so that it required refurbishment before using the accelerator.

Figure 3-4: DANTE accelerator with column removed for inspection. The low-energy section is to the left of the terminal and the high-energy section is to the right of the terminal.

In order to generate the high voltages required in a compact fashion, a modified version of the Cockroft Walton multiplier [94] is used in DANTE. The basic circuit works as follows: First, a low voltage (0-300V), high-frequency (30 kHz) signal is produced in a switching power supply. This signal is fed into an oil-filled transformer, which multiplies the voltage by a factor of 200. The signal then moves into the multiplier circuit, sometimes referred to as a "Jones and Waters" multiplier [95]. The multiplier consists of a two-sided "ladder" made of capacitors and diodes, topped by an inductor. As voltage is applied to the ladder, current moves through the diodes, charging each capacitor. Since current only moves in one way through the diodes, the ladder is charged and the voltage is increased at each step as well as being rectified into a DC voltage. A ladder with N steps will provide a final voltage of N times the
peak-to-peak AC input voltage from the transformer. A large (~1 H) inductor sits at the top of the ladder to provide impedance matching for the system capacitance.

While the switching power supply has varistors to protect the supply against feedback from arcs, the varistors can decay with repeated use, and a previous arc had overloaded a few varistors, causing them to burn and damage part of the power supply control board. The varistors and other burnt components were replaced to render the power supply operational again. In addition to damaging the power supply, the large arc destroyed the matching inductor and a few of the high-voltage diodes and capacitors in the multiplier circuit. The acceleration column was removed from the SF$_6$ tank (Figure 3-4), all of the diodes and capacitors were removed and inspected using a high voltage (10 kV) power supply, and the compromised components were replaced. Due to its large inductance and unique size/voltage requirements, the matching inductor was not able to be purchased directly, and had to be fabricated. Details of the inductor fabrication can be found in Appendix A. The oil-filled transformer was drained and inspected, but no damage was observed.

**Ion Source**

In addition to the high voltage acceleration system, repairs were required on the ion source, which had sustained significant damage after a loss of cooling accident. Resistive heating of the filament as well as heat from the source plasma itself necessitates a water cooling system for the ion source to keep components, particularly the Teflon insulators electrically separating different electrodes, from melting. During previous operation, the water cooling manifold had failed while the source was running, leading to melting of the insulators and deformation of the copper plasma and extraction electrodes. As a result of this damage, the decision was made to replace the source entirely with the spare ion source from a similar accelerator built by NSI. Because the dimensions of the replacement source were slightly different than the dimensions of the original source, it was necessary to reoptimize the source potentials.

An open-source 2D and 3D electrostatic code called IBSimu was used to simulate beam extraction and transport in the source and Einzel lens [96]. The code discretizes the simulation domain using the finite difference method and then solves the time independent Poisson equation and the time independent Vlasov equation using the biconjugate gradient stabilized method (BiCGSTAB) [97].

The general method of solving the problem is to first guess the electrostatic potential by simply solving for the Laplacian $\nabla^2 \phi = 0$ given the boundary conditions set by the geometry and potentials supplied by the user. The second step is to solve for the trajectories of the beam particles and calculate the space charge due to the beam (found by using the Vlasov equation) so that the Poisson equation ($\nabla^2 \phi = -\rho/\varepsilon_0$) can be solved for. The second step is repeated until the solution to the Poisson equation converges. In addition, IBSimu allows the user to input a prescribed magnetic field, which modifies the particle trajectories and the solution to the Vlasov equation.
Using a 2D axisymmetric CAD drawing of the source body, potentials were assigned to the different surfaces and beam trajectory and emittances were compared for different combinations of potentials in order to determine the optimum voltages for each electrode.

![Graphs and diagrams showing extraction and acceleration electrodes, and spatial beam profile.]

**Figure 3-5:** Beam emittances and spatial profile for the ion source optics. The negative hydrogen ions (red) are accelerated through the ion source while the electrons (yellow) are steered into the extraction electrode with a set of filtering magnets. The ion trajectories are slightly affected by the filter magnets (as seen by the offset in emittance plots from center) but this is corrected later in the Einzel lens.

As can be seen in Figure 3-5, the magnets in the extraction electrode are extremely effective in steering the electron beam into the side of the extraction electrode and out of the beam path. The position of the diverted electron beam in the simulation corresponds directly to visually-observed discoloration that has been observed on the extraction electrode, as seen in Figure 3-6.

After optimizing the front of the source, the Einzel lens was added in order to determine the correct focusing voltages. As can be seen from Figure 3-7, the Einzel lens focuses the beam by creating an electric potential structure which focuses the beam similar to an optical lens focusing light. By using a decelerating, and then accelerating field, the Einzel lens is able to focus the beam as well as accelerating it to the energy of the high-potential electrode of the lens. The negative, almost horizontal,
The slope of the emittance plot suggests that at the end of the Einzel lens, the beam is still slightly converging, but has almost reached a waist. An inspection of the beam profile shows a roughly circular beam profile with a radius of approximately 3 mm. The intensity of the beam profile plot shows a 1 mm radius bright spot in the beam, surrounded by a halo at 2.5 mm. By performing an optimization with the beam dynamics code, nominal operating parameters for the source and Einzel lens were found as summarized in Table 3.1.

<table>
<thead>
<tr>
<th>Source Parameter</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Plasma Electrode [V]</td>
<td>9.4</td>
</tr>
<tr>
<td>Extraction Electrode [kV]</td>
<td>1.07</td>
</tr>
<tr>
<td>Acceleration Electrode [kV]</td>
<td>8.52</td>
</tr>
<tr>
<td>Focus Electrode [kV]</td>
<td>1.55</td>
</tr>
</tbody>
</table>

Table 3.1: Optimal DANTE beam parameters

The final piece of the source is the filament. The filament is a 17.8 cm long, 2.4 mm diameter rod bent into a U shape (Figure 3-8). In order to increase electron emission, thoriated tungsten is used as the filament material. In order to "condition" the thoriated tungsten filament, Langmuir [98] recommends bringing the filament up to a temperature of 2600 K in order to reduce the thorium oxide in the filament to metallic thorium which spreads throughout the filament. However, since the thorium will evaporate from the surface at such high temperatures (thus reducing electron emission), the filament should be operated at a temperature between 2000 and 2100 K, to balance the surface evaporation of thorium with thermal diffusion of thorium to the surface.

While it is difficult to directly measure the temperature of the filament, the temperature can be estimated by using Ohm’s law. The effective resistance of the copper lines feeding the filament is estimated by measuring the resistance of the system while
Figure 3-7: Beam simulation through the whole Einzel lens. Note how the beam center has been moved almost entirely back to the center of the ion source by the focusing of the Einzel lens. Emittance and YZ beam profile plots taken at the exit of the Einzel lens.

cold and subtracting the tungsten resistance calculated from the geometry of the filament. Since the dimensions of the filament are precisely known, the resistivity of the filament can be calculated as:

\[
\frac{(V - IR_{Cu})A}{IL} = \rho_W
\]  
(3.3)

where \(V\) and \(I\) are the voltage and current measured at the filament power supply, \(R_{Cu}\) is the approximate resistance of the copper feed wires, and \(L\) and \(A\) are the length and cross sectional area of the tungsten filament. At high temperatures, the resistivity of tungsten is approximately linear with temperature [99]:

\[
\rho_W = 3.44 \times 10^{-10}T - 1.09 \times 10^{-7}
\]  
(3.4)

where \(\rho_W\) is in \(\Omega\)-m and \(T\) is in K. Since the copper lines do not heat up appreciably compared to the tungsten filament, the cold resistance of the copper is used, and the filament temperature for a filament with the above dimensions can approximately be
given as:

\[ T_{fil} \approx \frac{(V - 0.016I) \times 7.3 \times 10^4}{I} + 316 \]  \hspace{1cm} (3.5) 

with \( T \) in units of K. In this way, the approximate filament temperature can be monitored by observing the voltage and current from the filament power supply. As the high-density plastic insulators separating the filament leads from the metal source body begin to deform at 100 C, it is critically important to keep the outer filament leads at temperatures lower than 100 C, so remote thermocouple readers were installed on the filament leads to ensure the leads stay below this temperature. In practice, it was found that in addition to the internal water cooling system, the addition of a fan to circulate air around the filament leads ensured that the temperature did not get too high.

Figure 3-8: The filament is connected to two leads, insulated from ground, and the gas feedthrough is visible below the filament leads.

### 3.1.3 Beam energy calibration

Once the accelerator had been repaired, it was necessary to calibrate the beam energy. While the terminal voltage is measured by a generating voltmeter and can be used to approximate the beam energy if the charge state of the beam species is known, this measurement is approximate and must be calibrated. A common technique to calibrate the energy of electrostatic accelerators is to measure the products of nuclear reactions which have resonance energies. A commonly used reaction is the \( ^{27}\text{Al}(p, \gamma)^{28}\text{Si} \) reaction which has a resonance at 992 keV [100]. Unfortunately, this only gives one calibration point. In order to obtain multiple calibration gamma peaks, the \( ^{19}\text{F}(p, \gamma\alpha)^{16}\text{O} \) reaction with gamma emission peaks at 6.13, 6.92, and 7.12 MeV can be used [101]. The cross section for this reaction has many resonances at low incident proton energy and represents an ideal calibration source, as long as the target is thin enough that the beam will not slow down enough within the target to have interference with multiple resonances (see Table 3.2).
<table>
<thead>
<tr>
<th>Incident Proton Energy (keV)</th>
<th>Resonance Width (keV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>669</td>
<td>3.8</td>
</tr>
<tr>
<td>872</td>
<td>4.3</td>
</tr>
<tr>
<td>935</td>
<td>6.5</td>
</tr>
<tr>
<td>1138</td>
<td>3.0</td>
</tr>
</tbody>
</table>

Table 3.2: Location and width of $^{19}$F$(p, \gamma \alpha)^{16}$O resonance gamma peaks used to calibrate DANTE beam energy.

![Calibration Data](image)

Figure 3-9: DANTE energy calibration data and fit.

To calibrate the beam energy, the terminal was brought to approximately the location of the known resonance and the beam was centered on a thin (50 nm) LiF target. The beam energy was changed in small increments around the peak, and the 669 keV, 872 keV, 935 keV, and 1138 keV gamma peaks were recorded. Since the reaction products from the $^{19}$F$(p, \gamma \alpha)^{16}$O reaction are so high energy, there are no interfering background peaks, and faster measurements could be taken by simply integrating all counts above $\sim$5 MeV instead of just integrating under a peak. Using this method, the excitation curve (corresponding to the relative cross section) for the reaction can be obtained, and the resonance peak can be matched with the terminal voltage which produced it. By performing this procedure for the four peaks in Table 3.2, a calibration curve for the terminal voltage was obtained to correlate the measured terminal voltage on the generating voltmeter with the "true" terminal value deduced from the beam energy at resonance shown in Figure 3-9.
### 3.2 Accelerator Irradiations

A 10 m reel of 2G REBCO HTS coated conductor was obtained from SuperPower Inc. to be used in this study. The REBCO is 4 mm wide and has a 7.5% Zr doping fraction (in the form of BZO nanorods) to increase pinning efficiency. In order to allow effective ion irradiation of the samples, the REBCO was obtained without the usual 20 μm copper stabilizer that presents the outward surface of working tapes. This leaves a ~2 μm silver coating over the superconducting layer which is thin enough to allow a ~MeV proton beam to penetrate into the superconducting layer during accelerator irradiation. While the exclusion of the copper stabilizer makes critical current measurements more difficult, it does not affect the performance of the superconducting layer itself. Samples cut from a 3 m section of the tape reel were irradiated with the DANTE accelerator using a 1.2 MeV proton beam.

![Proton range](image1)

(a) Proton range

![Energy to primary recoil atoms](image2)

(b) Energy to primary recoil atoms

Figure 3-10: SRIM simulations for 1.2 MeV protons on silver-capped REBCO tape. These simulations show that for the samples being investigated, the beam particles are deposited far beneath the REBCO layer and the average energy to recoils in the REBCO layer is approximately constant.

While the primary-knock-on (PKA) energy spectrum of protons on REBCO is lower than that of neutrons in REBCO, protons have a much lower stopping power in REBCO than heavier ions and can be considered approximately monoenergetic in the superconducting layer. Monte Carlo calculations performed with SRIM [102] show that the beam will slow down 200 keV in the 2 μm silver cap layer and the average proton energy reaching the superconducting layer will be 1 MeV. SRIM simulations also showed that nearly all of the beam particles are deposited in the Hastelloy layer, far beneath the superconductor (Figure 3-10a) and that the average energy to recoils in the superconducting layer is approximately constant (Figure 3-10b). This is in contrast to heavier ions which have a strongly increasing energy to recoils deeper into the layer, effectively producing different damage in different depths of the superconductor. A comparison of primary recoil spectra for three different incident proton energies in 3-11 demonstrates the insensitivity of the recoil spectrum within a few
hundred keV incident proton energy.

Figure 3-11: Comparison of primary recoil energy spectra for 800 keV, 1 MeV, and 1.2 MeV proton beams on YBCO to illustrate the insensitivity of primary recoils to incident proton energy within a few hundred keV of the average proton energy 1 MeV in the REBCO. The spectra are overlaid for most of the energy range and only diverge at energies close to the beam energies themselves.

Recent ion irradiation work has shown that beam-induced drag can cause low-energy (low eV range) carbon atoms to be entrained in the primary beam and deposited on the target, leading to beam-induced carbon contamination of surfaces which in turn affects defect formation as carbon diffuses into the bulk material [103]. For the REBCO irradiation studies however, the existence of the 2 µm silver cap layer should obviate these concerns. In the study referenced above, the large increase in carbon concentration happened mostly in the first µm of the irradiated material, and by 2 µm, the irradiated carbon concentration was nearly identical to the unirradiated carbon concentration. In addition, the study referenced above was performed at a temperature of 748 K, 325 K higher than the highest temperature irradiation in this thesis. Thus, beam-induced carbon contamination in the REBCO layer was not assumed to be a big concern.

Effort was taken to ensure areal uniformity of irradiation over the entire sample. Critical current measured using the four-probe transport method will be limited by the most damaged region on the tape, so any irradiation “hot spots” caused by uneven beam coverage will result in artificially low critical current measurements. To ensure beam uniformity, the proton beam profile was first determined by performing intensity analysis of a CCD image of the beam on a gold-coated quartz window the same distance in beam drift space as the REBCO target holder in an adjacent beamline (see Figure 3-12). The beam focus was adjusted so that the beam spot size at >75% of peak intensity was large enough to cover the entire HTS target area. A custom image analysis code called BEATRICE was used to analyze the beam intensity on the window and provide vertical and horizontal beam intensity profiles shown in Figures 3-13a and 3-13b.
After a satisfactory beamspot was achieved the beam was steered onto the REBCO sample holder, where it passed through a set of collimators before impinging on the REBCO target (see Figure 3-14). The first collimator is slightly larger than the desired target outline and removes the heat load from the unused portions of the beam. The second collimator constructed from G-10 outlines the 6x4 mm irradiation area on the REBCO and has four copper pickups at the edge of each side of the opening to measure instantaneous beam current. The beam was centered on target by ensuring that the measured beam currents are the same on opposing sides of the rectangular collimator opening.

Fluence was determined by assuming the beam on target was approximately uniform (from image analysis above) and dividing the measured beam current on target by the beam charge and target area. To ensure accurate beam current collection, a suppression electrode was biased to a negative voltage to deflect secondary electron emission back to the target to avoid spurious current measurement due to ejected electrons. Operational experience showed that a suppression bias voltage of -200 V was sufficient to eliminate secondary electron effects.

In order to directly measure proton current on the REBCO sample, the sample must be isolated from electrical ground. Because most electrical insulators are also good thermal insulators, this leads to a situation in which the thin, thermally insulated REBCO samples can heat up quickly due to the power of the ion beam, in this case on the order of 0.4 watts for a 300 nA, 1.2 MeV beam. While 0.4 W is a negligible heat load on a sample with air convection cooling, the target was irradiated in vacuum and conduction through the sample holder was the only way to exhaust heat (radiation was negligible at the desired operating temperatures). In order to mitigate beam-related sample heating, the G-10 electrical insulator (with a thermal conductivity of 0.29 W/m-K [104]) was chosen to be as thin as possible (0.5 mm) to maximize heat transfer to the thermal sink behind the sample. Carbon tape was
Figure 3-13: The beam image in 3-12 was scaled from pixels to mm and vertical and horizontal slices of the beam profile were obtained. In each slice, a second scale was placed at 75% of the peak intensity in order to obtain an estimate of the beam width at 75% of full intensity. Note that the beam has been tuned so that the Gaussian peak is not symmetric in 2D, as the 6x4 mm collimator is wider in the horizontal direction.

used to mount the HTS tape to the G-10 insulator to facilitate good thermal contact. Sample temperature was recorded throughout each irradiation through the use of two type-K thermocouples, and instantaneous beam current to the target was limited to reduce the effects of beam heating.

### 3.2.1 High-temperature target stage

In order to investigate the effect of irradiation at elevated temperatures, a target stage was constructed to allow the HTS sample to be maintained at a constant temperature throughout the irradiation. The HTS target was mounted on a 2 inch diameter 0.5 inch thick aluminum cylinder, which was in turn connected to a 20 inch aluminum rod extending through a vacuum feedthrough. This large volume of aluminum acts as a heat sink and conduction path for the target and can also be cooled convectively outside of the vacuum chamber. The target assembly was modeled in COMSOL, shown in Figure 3-15. The target was found to reach steady state in approximately 10 seconds, a timescale much shorter than the shortest irradiations. “Worst case” heating conditions using a 1 W beam heat load (1 μA at 1 MeV) show that target temperature rises approximately 35 K in steady state beam operation. Based on the COMSOL simulations, a nominal beam current of 300 nA (~ 0.5 W beam load at 1.2 MeV) was chosen for the irradiations to minimize sample heating. Active heating
Figure 3-14: Image of HTS target mount. In Figure (a), the top disk is the first collimator, followed by the secondary electron suppression electrode, with the secondary G-10 collimator mount at the bottom. Figure (b) is a close-up of the target area with the collimator and suppression electrode removed, showing the four current pickups used to center the beam on target during operation.

was accomplished by using two 50 W cartridge heaters controlled by a PID. Sample temperature was measured using two type K thermocouples placed directly adjacent to the irradiated sample area. The thermocouples were read using a Pico TC-08 temperature logger. The TC-08 contains built-in cold junction compensation and can measure temperatures with an accuracy of ± 1.3 K. Using this method, sample temperature was able to be maintained from a range of 323 to 423 K with fluctuations of approximately ± 3 K due to source current fluctuations and PID response time.

Figure 3-15: Comsol target heating model displaying maximum target temperature rise (at steady state) for a 1 μA at 1 MeV beam heat load with a starting temperature of 300 K.
3.2.2 Cryogenically cooled target stage

In order to investigate the effect of cryogenically sample irradiations, a second target stage was constructed. A 6 W, two-stage Gifford-McMahon (GM) cryocooler [105] was modified to accept the aluminum target mount described above and fit into the beam chamber (see Figure 3-16). Through testing on a dummy sample, it was found that the cryocooler could maintain a sample temperature of approximately 80 K during irradiation with a 500 nA, 1.2 MeV proton beam. Thus, 80 K was used as the set point for cryogenic irradiations. As with the heated target stage, two type K thermocouples were used to measure sample temperature during irradiation. As 80 K was the minimum temperature achievable with a beam load, no PID control was used during cryogenic irradiations and beam parameters were tuned by hand to achieve as constant of a beam current as possible to minimize sample temperature excursions. As with the high-temperature irradiations, the temperature drift was approximately ±3 K for the cryogenic irradiations.

![Cryo-cooler Target Chamber Beamline](image)

Figure 3-16: HTS cryogenic irradiation chamber.

3.2.3 Data acquisition and control systems

A LabView control and data acquisition system was developed for the target chamber assembly. This system was used to control the active secondary electron suppression as well as monitor the target vacuum, sample temperature, and sample current. In
addition, temperature monitoring of the DANTE filament leads and source canister was implemented to ensure that no melting of the Teflon ion source insulators occurred. A software charge integrator was implemented to act as a check on the analog target current integrator in the DANTE control system. Relevant parameters (sample temperature, sample current, integrated sample charge, the presence of secondary electron suppression, and target chamber vacuum pressure) were recorded to file at half-second intervals. Example traces for temperature and pressure for one of the long cryogenic irradiations are shown in Figure 3-17 to demonstrate the stability of the sample operating conditions during irradiation.

Figure 3-17: Traces of target temperature (a) and chamber pressure (b) vs. time for one of the cryogenic irradiations. Current is also plotted in both figures to indicate when the beam is on target. Although the beam current changes due to changing plasma conditions in the ion source, the target temperature and pressure are maintained at approximately constant levels throughout the irradiation.
Chapter 4

REBCO Characterization

For fusion magnets, the practical quantity of interest is the maximum current that can be sent through a superconducting tape of fixed cross section. The higher the current, the less tape required to provide the same magnetic field and the more room available for structure (see Eq. 2.6). If the strength of the magnet is limited by the structural self-forces on the magnet (which is the case for compact reactors), the critical current of the superconducting material effectively sets the maximum magnetic field which can be provided by the superconducting magnet. As such, the critical current has been used in this study as the metric of “performance” of the superconductor.

In order to obtain the critical current and n-values (an indication of the superconducting-normal transition sharpness) of the proton-irradiated superconductors, the four-probe transport method was utilized. Preliminary $I_c$ analysis of accelerator-irradiated samples was performed at MIT using liquid nitrogen, while later analysis was performed at the Robinson Research Institute in New Zealand using a gas-cooled transport current measurement system to perform high-fidelity $I_c$ analysis of the irradiated samples.

4.1 $I_c$ measurement method

There are two primary ways to measure the critical current of a sample. The first, called the magnetization method, offers increased resolution but relies on the interpretation of a theoretical model [106] to determine the current and can diverge from the “true value” of critical current at higher magnetic fields [107]. The second, known as the four-probe transport method [42], involves directly applying a current to the sample and measuring the voltage across the superconducting region of interest, schematically shown in Figure 4-1. This method gives a measure of the practically achievable critical current for tape being used in a superconducting fusion magnet. While in the superconducting state, the superconductor has no resistance, and therefore due to Ohm's law ($V = IR$), no voltage will be measured across the region.
However, once the superconducting state is lost, the material will become resistive, and a sharp increase in voltage can be measured (see Figure 4-2).

![Schematic of four-probe critical current measurement.](image)

**Figure 4-1:** Schematic of four-probe critical current measurement. Orange represents the superconducting REBCO tape, while the other components (gray and black) are normal conductors. A power supply is connected to both ends of the superconductor (while it is cold) to provide current. Voltage is measured across a known distance as current is increased and an electric field criterion (typically a rise of 1 \(\mu\)V/cm) is used to determine the transition between the superconducting and normal state.

A typical threshold electric field used to determine critical current in REBCO using the transport method is 1 \(\mu\)V/cm \([6, 31, 32]\) and was used for this work. In a practical measurement, there will always be some voltage floor caused by thermal fluctuations within the wires connecting the superconducting sample to the nanovoltmeter. In order to minimize this noise, very fine magnet wire is used for the voltage measurement leads, and the connection to the nanovoltmeter is made well outside of the cryogenic measurement area to reduce large thermal fluctuations.

### 4.1.1 n-value

In addition to the critical current, the n-value of the irradiated superconductors was measured. As mentioned above, the transition between the superconducting and normal state in the four-probe transport method is characterized by a sharp voltage increase. This increase can be modeled as a power fit:

\[
\frac{V}{V_c} = \left(\frac{I}{I_c}\right)^n
\]  

(4.1)

where \(V_c\) represents the voltage threshold criterion (1 \(\mu\)V/cm), \(I_c\) represents the critical current defined as the current through the sample when \(V = V_c\), and \(n\) is the exponent which characterizes the sharpness of the voltage increase. The n-value is typically used as a measure of superconducting quality, i.e. the higher n-value, the
Figure 4-2: Example $I_c$ transport measurement for a sample with 0.4 cm between voltage taps. Note that the fit bounds have been adjusted to exclude the noisy baseline region near zero current as well as the region at high current where the superconductor starts to become fully normal. The n-value for this measurement was 18.7.

higher quality the superconductor, as it can operate very close to its transition temperature. Thus the n-value is of practical use to the SC magnet designer. While the physical mechanism causing changes in n-values for low-temperature superconductors has been determined to be current inhomogeneities in the critical current distribution [108], the mechanisms behind n-value changes for HTS are not fully understood. Recent work measuring irradiated superconductors has suggested that the n-value in HTS is affected by pinning mechanisms and forces [34].

4.2 Critical current analysis with the SuperCurrent measurement system

In order to achieve a large scan of high-fidelity measurements, the accelerator-irradiated samples were brought to the Robinson Research Institute (RRI) for analysis with their automated SuperCurrent measurement system [109] (see Figure 4-3). The SuperCurrent can sweep through a desired set of fields (from 0-8 T), temperatures (15-90 K), and field angles (0-240 degrees), obtaining the V-I transport curves at each combination. Operating in this fashion, the RRI SuperCurrent was used to collect approximately 18,000 $I_c$ measurements of the irradiated and control samples.
4.2.1 The importance of full angular resolution

Until recently, only two field orientations were important when measuring the critical current in REBCO – the 0 degree (field perpendicular to tape, sometimes denoted “parallel to c” because the crystal c-axis is always perpendicular to the tape face) and 90 degree (field parallel to tape) orientations. Up until recently, almost all REBCO tapes displayed similar angular behavior: a field angle of 90 degrees corresponded to a peak in $I_c$, with $I_c$ decreasing on either side of the 90 degree peak to a minimum value at 0 degrees (see Fig. 4-4). In order to characterize tapes for use in engineered structures such as fusion magnets using transposed tapes, it was only necessary to know the minimum $I_c$, which occurred at fields perpendicular to the tape.

However, recent work [65] has emphasized the breakdown of the above assumption with the increasing use of artificial dopants such as BZO nanorods, which act as correlated pinning centers and improve critical current at angles of 0 and 180 degrees but not intermediate angles, leaving the true $I_c$ minimum somewhere between 0 and 90 degrees. This underscores the necessity of obtaining full angularly-resolved scans of $I_c$ for three reasons. The first, obvious reason is that it is no longer sufficient to know the $I_c$ value at approximately 0 degrees when evaluating superconducting wire for use in magnets, as the 0 degree $I_c$ might not be the actual minimum $I_c$. The second reason is that, as shown in Figure 4-5, the angular dependence of the critical current can illuminate whether artificially-induced pinning centers (whether introduced through irradiation or in the REBCO fabrication) display a preferential
Figure 4-4: Example of a normal (undoped) REBCO critical current angular dependence, with a peak at 90 degrees and a minimum close to 0 and 180 degrees.

direction in relation to an applied field. The final reason is that the field-to-tape angle in a large fusion coil will sweep through all angles since it is likely that the conductor will use twisted tape geometry and the poloidal to toroidal field ratio varies during tokamak operation.

Figure 4-5: Example of angular dependence caused by artificial correlated pinning (in this case due to BZO nanorod inclusions). Note that the 0 and 180 degree currents (typically the minimum current) are almost as high as the 90 degree current.

In order to investigate the effects of radiation on correlated pinning, fully angular-resolved scans were performed on the ion-irradiated samples.

4.2.2 Preparing the samples

In order to analyze the REBCO samples in the SuperCurrent measurement device, the samples had to be removed from the G10 irradiation stage coupons, where they
had been affixed with carbon tape. Especially in samples which had experienced heating, the REBCO tapes were firmly stuck to the carbon tape and had to be removed carefully to prevent mechanical damage. First, the entire irradiation stage was soaked in acetone for approximately 10 minutes to loosen up the carbon tape. Then a sharpened wooden stick was used to gently push the REBCO sample off of the carbon tape, taking care not to bend the REBCO sample and cause mechanical damage (see Figure 4-6). Residual carbon tape was cleaned off using acetone.

Figure 4-6: Sample removal from the G10 irradiation stage coupon. Care was taken to not bend or scratch the samples during removal, which is why only plastic and wooden tools were used for this process.

Once the REBCO samples had been removed from the irradiation stage and cleaned, the irradiated area was patterned into a 4 mm long x 0.5 mm wide bridge. The purpose of patterning the samples was twofold. First, the small bridge pattern lowered the applied current required to reach $I_c$, allowing investigation of low-temperature regimes where $J_c$ is high. Second, the standard bridge pattern ensured reproducibility between sample measurement cross sections. To pattern the samples, the irradiation area was marked and a photomask was used to apply the bridge pattern to the sample. The sample was then immersed in a 10:3:1 mixture of water, hydrogen peroxide, and ammonia to remove the silver overlayer from the unmasked portions of the sample. This was followed by an immersion in an ethylenediaminetetraacetic acid (EDTA) solution to remove the YBCO underneath the etched silver areas. The processed samples were then cleaned one more time with acetone to remove the photo mask solution. An advantage of the particular bridge pattern shown in Figure 4-7 is that each “pad” charges to the potential at the end of the bridge, allowing voltage leads to be soldered anywhere on the pad while still allowing voltage measurements to be performed across the full, precisely known length of the bridge.
Figure 4-7: Sample preparation process. Figure a shows the sample after the application of the photolithography mask. Figure b shows the sample after silver etching (the exposed YBCO layer is black). Figure c shows the sample after the YBCO etching (the exposed buffer layer is a reflective green color). The final bridge pattern is shown in Figure d.

4.2.3 Mounting the samples

The SuperCurrent system utilizes a warm-bore magnet, allowing sample installation to be accomplished by the use of a ~1 m sample rod which can be warmed and pulled out of the measurement system without warming the magnet. Once samples had been patterned according to the above procedure, the ends to be connected to the current supply were soldered to two large silver foils using InBi solder at a temperature of 140°C. Large pads on the sample rod were cleaned and tinned, and the silver foils could then be soldered to the large current contacts at a higher temperature of 250°C without the risk of heating the superconducting sample. Once the sample had been soldered to the end of the sample rod, a small amount of Apiezon N thermal contact grease was applied to the bridge area, and a Cernox temperature sensor affixed to a thin piece of sapphire (electrically insulating but thermally conductive) was pressed against the sample. Finally, the voltage taps were soldered on to the sample pads using a soldering iron temperature of 140°C (see Figure 4-8).
4.3 Data collection and analysis

Data collection for the critical current measurements consisted of custom acquisition software from RRI/HTS-110 Inc. which recorded values of voltage across and current through the sample as well as the sample operating conditions of field, temperature, and field angle (see Figure 4-9). Critical currents were determined by fitting the data to the power law described in Eq. 4.1 using a non-linear fit with $n$ and $I_c$ as free parameters. Due to the large amount of measurements, the MDSPlus framework [110] was used to store the data in a central repository and create a SQLite database with metadata (such as sample irradiation conditions). The combination of the SQLite database and MDSPlus trees was then used to write analysis programs which could quickly compare datasets at different measurement and irradiation conditions.

4.3.1 Repeatability of measurements and error analysis

Although error bars are generally not reported for critical current measurements, an attempt was made to quantify measurement uncertainty. First, repeat measurements of the same sample were performed to establish measurement uncertainty of the SuperCurrent device. Although it would be infeasible to take multiple repeat measurements to calculate error bars individually for each $I_c$ measurement, a dedicated scan was performed on one sample multiple times (without remounting), as shown in Figure 4-10. Three full angular resolved $I_c$ scans were performed for each of the different field and temperature conditions shown in the plot.

The three values of $J_c$ for each measurement condition (field, temperature, and field
angle) were averaged, and the standard deviation of the group was computed. The calculated standard deviations were relatively consistent across all angles, fields, and temperatures, with the exception of the 7T, 30K measurements around 90 degrees. The explanation for this is most likely due to the high Lorentz forces on the sample at the high current and field bending the sample so that it is not flat with the Hall sensor mounted inside the sample rod. Due to the sharp peak in $I_c$ around 90 degrees, even a small discrepancy between the measured Hall angle the actual angle of the sample with the field could cause a large discrepancy between two measurements. Unfortunately, it was impossible to measure sample deflection during a measurement, so the only way to correct for this error is to compare full angular scans between measurements and note when the 90 degree peaks are shifted. In order to establish error bars for the critical current measurements, the standard deviations in Figure 4-10 were averaged to yield a global standard deviation of 1.3%, which was applied to the data analysis in Chapter 5.

Another possible source of error is due to manufacturing defects in the REBCO samples themselves. Because transport critical current measurements involve soldering, it was impractical to measure the same sample before and after irradiation. Thus, when comparing unirradiated and irradiated samples, two separate physical samples had to be measured, introducing the possibility of error due to pre-existing differences in $I_c$ of the irradiated sample when compared to the unirradiated control. This error is extremely difficult to quantify because while manufacturing errors are unpredictable, they can often follow a pattern due to a glitch in the manufacturing process. For example, a bad bearing in a tape spool on a manufacturing production line could introduce small, periodic degradations in tape $I_c$ with a frequency component corresponding to the revolution period of the payout. In fact, manufacturers have recently begun to perform Fourier analysis on high-fidelity $I_c$ measurements of production
In order to reduce sample variability due to manufacturing processes, all samples analyzed in this thesis were taken from a continuous 2.5 meter length to ensure that the processing conditions were as similar as possible. To remove the effect of remaining variations, a full TapeStar characterization of the experimental tape spool was obtained from SuperPower (see Figure 4-11). Every time a sample was cut from the spool the length was recorded to ensure that the position of each sample with respect to the TapeStar measurement was known. The control sample was used as the “standard” for critical current, and critical current values for all other samples were scaled relative to the control (which was at position 49680 cm in the spool).
Other sources of systematic error could come from variable irradiation conditions. The most likely candidate is dose rate, as has been discussed extensively in the literature for fission material irradiations [112, 113]. However, much of the discussion concerning dose rate concerns metals, not ceramics like REBCO. While best efforts have been put forth to keep dose rate (i.e., beam current on target) constant over each irradiation, negative ion sources are known for having variable beam currents and it was difficult to maintain a constant current on target within a factor of 50%. To investigate the question of whether dose rate plays a role in radiation damage of REBCO, a sample was irradiated to a fluence of $1.0 \times 10^{16}$ p/cm$^2$ at a beam current approximately three times lower than normal. This sample was compared to another sample irradiated to the same fluence but at the normal dose rate corresponding to a beam current of $\sim 300$ nA. Comparisons of critical current were carried out for a high-temperature, low-field (77 K, 1 T) measurement and a low-temperature, high-field (30 K, 5 T) measurement. As can be seen in Figure 4-12, the critical currents of both samples are very similar, especially in the high-field, low-temperature region of interest for fusion magnets. Due to the agreement between the two samples irradiated at a factor of 3x different flux, minor variations in beam current during the normal sample irradiations were assumed to not introduce significant changes in measured $I_c$. It should be noted that in order to achieve fluences comparable to the “lifetime” dose received in a fusion magnet, the damage rate for the irradiations in this thesis was several orders of magnitude greater than would actually be experienced by REBCO in a fusion magnet. Nevertheless, for the purpose of the comparisons between irradiation temperatures investigated in this thesis, moderate changes in dose rate do not appear to introduce systematic error.

![Figure 4-12: Comparison of critical current measurements between a normal beam flux and 3x reduced beam flux at a given fluence. The critical current measurements are similar, leading to the conclusion that dose rate, at the damage rates used in this study, does not play a large role in $I_c$ degradation due to irradiation.](image-url)

Another possible source of error comes from the effects of target heating. At high temperatures, oxygen will diffuse out of the REBCO matrix, lowering $T_c$ and eventually causing $I_c$ degradation. While tape manufacturers publish guidelines for maximum
safe temperatures to solder at, it is generally assumed that these temperature ranges are for soldering, which is a short duration procedure. SuperPower recommends a solder temperature of 195°C (468 K) and to not exceed 240°C (513 K). Since efforts were taken to irradiate samples at a uniform dose rate, the high fluence, high-temperature irradiations remained at 423 K for long periods of time, up to 80 minutes. In order to test whether simply keeping samples at an elevated temperature induces $I_c/T_c$ degradation, an unirradiated sample was brought up to 423 K in vacuum using the heated target sample mount for 80 minutes. Angularly-resolved measurements of $I_c$ were then carried out at the two temperature/field conditions and compared with the unirradiated, unheated control sample.

Figure 4-13: Comparison of critical current measurements between an unirradiated sample heated to 423 K for 80 minutes and the unirradiated, unheated control. An observation of the two plots shows that sample heating by itself does not lead to significant $I_c$ degradation at angles besides 90 degrees.

As shown in Figure 4-13, a comparison of the heated control and unheated control in the high-temperature/low-field and high-field/low-temperature measurement regimes shows minimal difference. Interestingly, there is a real, small decrease in both measurement regimes near the 90 degree field angle, possibly a result of diffusion-related disorder in the Cu-O planes leading to a decrease in correlated pinning.
Chapter 5

Critical Current Analysis of Radiation Damaged REBCO

Using 1.2 MeV protons provided by the DANTE accelerator, REBCO samples were irradiated to four different fluences ($1 \times 10^{15} \text{ p/cm}^2$, $5 \times 10^{15} \text{ p/cm}^2$, $1 \times 10^{16} \text{ p/cm}^2$, and $5 \times 10^{16} \text{ p/cm}^2$) at three different irradiation temperatures (80 K, 323 K, and 423 K). The highest fluence value was chosen to approximately match the DPA (0.003) at which previous studies have observed $J_c$ degradation due to neutron irradiation. The RRI SuperCurrent system was subsequently used to analyze critical current in the irradiated samples. The primary question this thesis seeks to address is whether irradiation temperature is important when investigating $J_c$ degradation in REBCO due to irradiation.

![Figure 5-1: Critical current density (at 5 T, 30 K) of samples irradiated at different temperatures to fluences of $1 \times 10^{16} \text{ p/cm}^2$ and $5 \times 10^{16} \text{ p/cm}^2$. Lines are drawn between data points to guide the eye. $J_c$ degradation shows a clear dependence on irradiation temperature at the higher fluence.](image)

The answer to this question is that irradiation temperature unequivocally plays a role...
in the $J_c$ degradation induced during irradiation, and the subsequent impact on $J_c$, as can be seen in Figure 5-1 displaying the critical current density of samples irradiated at different temperatures to fluences of $1 \times 10^{16}$ p/cm$^2$ and $5 \times 10^{16}$ p/cm$^2$. At measurement conditions relevant to a compact, high-field fusion reactor (5 T, 30 K), the irradiation temperature is shown to degrade the minimum $J_c$ by approximately a factor of 2 between the 80 K and 423 K irradiation at the higher fluence. This result has significant implications for fusion magnets, as all REBCO irradiations to determine the lifetime of the superconductor in a fusion environment have been performed at temperatures between 323 K and 353 K [6, 114, 115].

![Figure 5-2: Selection of plots illustrating irradiation temperature effect on REBCO $J_c$ degradation due to proton irradiation at various measurement fields and temperatures, with lines connecting data points to guide the eye. There appears a transition between $1 \times 10^{16}$ p/cm$^2$ and $5 \times 10^{16}$ p/cm$^2$ from a regime which is minimally affected by irradiation temperature to a regime in which there is a large difference between cryogenic and high temperature irradiation conditions.](image-url)
the irradiation will occur at $T < 80$ K. Since Figure 5-2 clearly establishes the importance of irradiation temperature for proton irradiations, the next logical question is whether these results are transferable to neutron irradiations. In order to determine whether the results in this paper could be relevant to neutron irradiations, it is important to understand the mechanisms behind the difference in $J_c$ degradation at different irradiation temperatures.

As described in Chapter 2, the effect of radiation on the $J_c$ of REBCO is a mixture of damage and enhancement mechanisms, with each of these mechanisms having different dependencies on fluence and operating regime of the tape. The relative contributions of these mechanisms is responsible for the variability of the $J_c$ degradations in Figure 5-2, and the following sections in this chapter will attempt to explain the features in this figure through different analyses of the critical current vs. field, temperature, and field angle data.

Before investigating the mechanisms behind enhancement or degradation of $J_c$ due to irradiation temperature, it is important to point out a key feature of Figure 5-2. The relative effect of irradiation temperature on $J_c$ degradation is noticeably larger for the larger fluence. At a fluence of $1 \times 10^{16}$ p/cm$^2$ and measurement temperature of 50 K, there is little difference in degradation between the two higher temperature irradiations, whereas for 20 K and 30 K operating temperatures, the 423 K irradiation has slightly less degradation than the 323 K irradiation. At the higher fluence, higher irradiation temperature always leads to a larger degradation in $J_c$.

### 5.1 Critical temperature modifications

It is well established [5] that a reduction in critical temperature will lead to a degradation of critical current density. As such, it is instructive to investigate the changes of critical temperature in the different irradiated samples. In order to conduct these measurements, scans of critical current vs. temperature were obtained and fit using the Ginzburg Landau theoretical dependence described in Eq. 2.14. An example $T_c$ fit is shown in Figure 5-3.

Critical temperatures were calculated for all irradiated samples, and are shown in Figure 5-4. The first noticeable trend is that (as expected) the critical temperature decreases as the irradiation fluence increases. Unexpectedly, for all three fluences the critical temperature appears to have a weak to nonexistent dependence on irradiation temperature. There is a clear drop in $T_c$ between $1 \times 10^{16}$ p/cm$^2$ and $5 \times 10^{16}$ p/cm$^2$ fluences, which is consistent with the clear break in $J_c$ degradation versus irradiation temperature shown in Figure 5-2, and thus suggests the $T_c$ effect is at least correlated to the $J_c$ degradation. Previous studies of low-temperature REBCO irradiations [73] [116] have asserted that between 20K and 300K, the irradiation temperature plays a small, if not negligible role in $T_c$ degradation. The results in Figure 5-4 indicate that while $T_c$ does not vary strongly with $T_{irr}$, there is a measurable difference between $T_c$
values for different irradiation temperatures at lower fluences.

Figure 5-3: Zero-field $T_c$ fit on the pristine control sample.

Figure 5-4: Critical temperature vs. irradiation temperature. Note that the zero of the y axis has been suppressed to highlight the changes between fluences.

Comparisons with the annealed sample help explain the behavior in Figure 5-4. In Figure 5-5, the irradiations to a fluence of $1.0 \times 10^{16}$ are re-plotted from Figure 5-4. In addition we show the $T_c$ values for the same fluence and irradiation temperature, but with a post-irradiation anneal. This sample was annealed after irradiation for 8 hours at 423 K, and the $T_c$ increased, recovering about half of the 1.2 K drop in $T_c$ from the non-irradiated sample and has a $T_c$ value consistent with the higher temperature irradiations. The partial recovery of $T_c$ after sample annealing suggests that at higher temperatures, recombination of Frenkel pairs proceeds fast enough to partially reverse the degradation of $T_c$. Overall, and particularly for the case of most
Figure 5-5: Critical temperature vs. irradiation temperature for a fluence of $1 \times 10^{16}$ p/cm$^2$. The annealed sample recovered to approximate the same critical temperature as both higher-temperature irradiations.

interest at high fluence, we conclude that $T_c$ degradation is not the primary driver for the dependence on irradiation temperature in Figures 5-1 and 5-2.

5.2 Differentiating strong and weak pinning regions

For the purposes of the analysis in the following sections, it is useful to break the $J_c$ measurement parameter space into two broad regimes – strong pinning and weak pinning. As described in Chapter 2, strong pinning sites distort the flux line lattice itself and are generally very stable against thermal lattice vibrations, while weak pinning sites act collectively to preserve the shape of the flux lattice and are more prone to being unstable to thermal vibrations. Thus, strong pins are more effective in conditions of high temperature and low field, whereas weak pinning sites are more effective at low temperature and high fields. One way to characterize these regions is by analyzing the variation of $\log(J_c)$ with $T$. The critical current density dependence on weak pinning has been shown to follow the relationship [64]:

$$J_{c,w} \approx J_{0,w} \times \exp \left[ - \left( \frac{T}{T_{0,w}} \right) \right] \quad (5.1)$$

where $J_{0,w}$ and $T_{0,w}$ are fit parameters proportional to the critical current density and pinning barrier energy at zero temperature (i.e. without thermal fluctuations leading
to flux creep and thermally activated depinning). It is important to point out that due to the extremely large parameter space over all possible field, temperature, and field angle combinations possible, the measurements performed on the SuperCurrent system were prioritized first with regards to angle, then field, then temperature. Thus, high resolution scans of $J_c$ with $T$ were not performed at many fields besides the self-field case used to determine $T_c$ and data at other fields was limited. Nevertheless, Equation 5.1 can be used to roughly approximate regions of the data. If the $J_c$ vs $T$ trend fits well to Equation 5.1 it is deduced that we are in the weak pinning regime, and where the data trend deviates from Equation 5.1, as $T$ increases, then this is identified as the transition temperature into the strong pinning regime.

Figure 5-6 compares the $J_c$ dependences with temperature for several fields (field oriented perpendicular to the tape) in the unirradiated control sample. Dotted vertical lines were plotted to guide the eye to the point where the data deviates from the fit to Eq. 5.1 by more than 5%. At zero field, the transition temperature between strong and weak pinning occurs at approximately 64 K and steadily decreases as the applied field increases, ending up at ~52 K for $B = 7$ T. While the poor resolution of temperature points in the higher field data means that the true transition temperature could be higher than indicated, the plotted result can be used as an approximate transition temperature.

Figure 5-6: Comparison of measured $J_c$ to fit with temperature in Eq. 5.1 (dotted line). Vertical dashed lines indicate the approximate transition temperature between weak and strong pinning regimes.

Figure 5-7 compares the $J_c$ dependences with temperature for several fields (field oriented at 0 degrees) in the samples irradiated to the highest fluence of $5 \times 10^{16}$ p/cm$^2$ at temperatures of 80 K and 423 K. As with Figure 5-6, dotted vertical lines were plotted to guide the eye to the point where the data deviates from the fit to Eq. 5.1 by more than 5%. Unfortunately the sample irradiated at 80 K mechanically degraded part of the way through the $J_c$ measurement and the $0$ T and $7$ T data were not obtained.
Figure 5-7: Comparison of measured $J_c$ to fit with temperature in Eq. 5.1 (dotted line). Vertical dashed lines indicate the approximate transition temperature between weak and strong pinning regimes.

The sample irradiated at 423 K (Figure 5-7 (b)) displays the same trend of decreasing boundary temperature with increasing field as the unirradiated sample, although all of the boundary temperatures are shifted downwards approximately 10 K from Figure 5-6. The most important feature to note is that the transition temperature in the 5 T case is $\sim$ 48 K for both 80 K and 423 K irradiation temperature. Therefore this suggests that fluence, rather than irradiation temperature, is the primary driver for lowering the weak-to-strong pinning transition.

To shed further light on the mechanism behind the shift in the transition temperature, the intermediate fluence ($1 \times 10^{16} \text{ p/cm}^2$) samples irradiated at 80 K and 423 K are plotted in Figure 5-8. The lower fluence irradiations have transition temperatures at values in between those seen at high fluence and unirradiated samples, which confirms that fluence, rather than irradiation temperature, seems to have most significant effect on the transition temperature.

For the range of measurement fields in this study there is clearly a region of operating temperature below $\sim$40 K that is always dominated by weak pinning and a clear region above $\sim$ 65 K always dominated by strong pinning. In the following section we will use this determination to distinguish behavior in one of the two regimes. The range in between these two temperatures is more complicated and appears to depend on the level of irradiation fluence and applied field. Higher fluence and higher applied fields both have the effect of pushing the crossover temperature to lower values. Due to the low resolution of the data, it is difficult to draw strong conclusions about the effect of irradiation temperature on the pinning regimes although it appears that the transition temperature shifts more strongly as a function of fluence than irradiation temperature.
Figure 5-8: Comparison of measured $J_c$ to fit with temperature in Eq. 5.1 (dotted line). The region where the log($J_c$) data is linear with temperature (thus matching the dotted fit line) is where weak pinning is active and the region where the data diverges from the fit indicates a transition from weak to strong pinning.

5.3 $J_c$ vs. $\theta$ comparisons

The main group of high-resolution measurements performed at RRI were high-fidelity angularly-resolved $J_c$ measurements performed at several different temperature and field combinations. The following sets of figures will investigate the effect of radiation fluence and temperature on the angular $J_c$ dependence. Each sample being analyzed has been compared to the unirradiated control sample to establish the degree of enhancement or degradation in $J_c$. In order to investigate the angular $J_c$ changes in both the strong and weak pinning regimes, two cases were compared for each sample. Based on the results of the previous section, the strong pinning condition was chosen to be 77 K, 1 T, and the weak pinning region was chosen to be 30 K, 5 T. It is important to note that the same behavior in the weak pinning regime was observed down to temperatures of 15 K (as expected), but due to the high measurement currents involved it was not possible to obtain 15 K measurements for all irradiated samples so 30 K is used as a baseline of comparison.

The first set of figures compares a range of increasing fluences at an irradiation temperature of 80 K. For each figure, the left plot is the strong pinning regime condition (1 T, 70 K), and the right plot is the weak pinning condition (5 T, 30 K). In Figure 5-9, the critical current density of the irradiated sample at low fluence increases approximately uniformly over the entire range of angles and in both pinning regimes suggesting the inclusion of effective pinning sites in both regimes. As fluence is increased to the intermediate ($1 \times 10^{16}$ p/cm$^2$) fluence in Figure 5-10 and high ($5 \times 10^{16}$ p/cm$^2$) fluence in Figure 5-11, $J_c$ drops across all angles in the strong pinning regime. The $J_c$ behavior in the weak pinning regime is more complex. As fluence is increased
Figure 5-9: Critical current density vs. measurement angle for 80 K, $5 \times 10^{15}$ p/cm$^2$ irradiated sample as compared to the unirradiated sample, in both strong (left) and weak (right) pinning regimes.

Figure 5-10: Critical current density vs. measurement angle for 80 K, $1 \times 10^{16}$ p/cm$^2$ irradiated sample as compared to the unirradiated sample, in both strong (left) and weak (right) pinning regimes.

Figure 5-11: Critical current density vs. measurement angle for 80 K, $5 \times 10^{16}$ p/cm$^2$ irradiated sample as compared to the unirradiated sample, in both strong (left) and weak (right) pinning regimes.
Figure 5-12: Critical current density vs. measurement angle for 423 K, $5 \times 10^{15}$ p/cm$^2$ irradiated sample as compared to the unirradiated sample, in both strong (left) and weak (right) pinning regimes.

Figure 5-13: Critical current density vs. measurement angle for 423 K, $1 \times 10^{16}$ p/cm$^2$ irradiated sample as compared to the unirradiated sample, in both strong (left) and weak (right) pinning regimes.

Figure 5-14: Critical current density vs. measurement angle for 423 K, $5 \times 10^{16}$ p/cm$^2$ irradiated sample as compared to the unirradiated sample, in both strong (left) and weak (right) pinning regimes.
from $1 \times 10^{16} \text{ p/cm}^2$ to $5 \times 10^{16} \text{ p/cm}^2$, $J_c$ at 90 degrees continues to drop. At 0 degrees, however, $J_c$ remains virtually unchanged, and the minimum $J_c$ in the region between 0 and 90 degrees actually increases with fluence. This strongly suggests the addition of coherent weak pinning centers as the fluence is increased, but the destruction of the strong correlated pinning from the Cu-O chain layers.

The second set of figures (Figures 5-12 to 5-14) compares a range of increasing fluences at the higher irradiation temperature of 423 K. In contrast to the irradiations performed at 80 K, none of the irradiations produced $J_c$ enhancement for the strong or weak pinning regimes. The decreases in $J_c$ are consistent across all irradiations for measurements performed in the strong regime, with increasing relative $J_c$ degradation at higher fluences. For the first two fluences, this degradation is more or less constant in angle, although for the highest fluence irradiation (Figure 5-14) the 90 degree peak appears to almost disappear completely. It should be noted that there was no 77 K measurement data for the strong pinning regime because $I_c$ was too small to be measured, so 50 K measurements were used instead. In the weak pinning regime, $J_c$ also degrades increasingly with higher fluences, although this effect is much more pronounced for the 90 degree peak area compared to other angles.

### 5.4 $J_c$ vs. B comparisons

A common way to study the effects of pinning (in the weak pinning regime) for fields with an angle of 0 degrees is to fit the dependence of $J_c$ vs. B with a power law of the form $J_c \propto B^{-\alpha}$ above fields of 3 T [117]. A higher value of $\alpha$ corresponds to a higher sensitivity of $J_c$ to the applied magnetic field (i.e. the $J_c$ degrades more rapidly with increasing B), implying less efficient flux pinning. Figure 5-15 shows the field dependencies of $J_c$ for the unirradiated control sample, with $\alpha$ values of approximately 0.65, consistent with previously reported values for unirradiated tape with BZO nanorod dopants [64].

The first set of B-field dependencies in Figure 5-16 for an irradiation temperature of 80 K shows the decrease of $\alpha$ with increasing fluence, suggesting further evidence for the creation of effective weak coherent pinning centers being introduced with irradiation at this temperature. The decrease of $\alpha$ at lower operating temperatures suggests small scale-size defects which would be more effective pinning sites as $\xi$ decreases with T. From a practical view, a lower $\alpha$ is highly attractive because it flattens the $J_c$ vs. B curve and improves tape viability at high absolute magnetic fields attractive for compact tokamak reactors. For example consider the case shown in Figure 5-11 (right) and Figure 5-16 (c) with cryogenic irradiation at high fluence, at high field, sub-cooled operating conditions. Not only does the 0 degree $J_c$ remain as high as the pristine sample (and the minimum $J_c$ over the angular range is actually increased), but the $\alpha$ has been decreased from $\sim0.68$ to 0.5. Thus the expected decrease in $J_c$ from 7.5 T (the highest B value obtained in this work) to $\sim22$ T (peak field at coil
Figure 5-15: $J_c$ vs. B for control sample with fitted power law $\alpha$ values. Error is represented as a shaded region.

Figure 5-16: $J_c$ vs. B curves for 80K irradiations with calculated power law $\alpha$ values. Error is represented as a shaded region.

for ARC) implies a decrease of only $(7.5/22)^{0.5} \sim 0.58$ in $J_c$. These results are highly encouraging when viewed in the context of lifetime survivability of REBCO tapes to be used in fusion magnets.

The second set of B-field dependencies in Figure 5-17 for an irradiation temperature of 423 K also shows the decrease of $\alpha$ with increasing fluence and decreasing operating temperature, suggesting that small, effective weak pinning sites are also being produced at this irradiation temperature. However, this decrease in $\alpha$ is smaller and also accompanied by a decrease in absolute $J_c$, unlike the irradiations at 80 K. The combination of these results implies that the higher-temperature irradiations have less of an effect at suppressing the creation of pinning sites as opposed to amplifying the amount of damage done to the superconductor by irradiation, although the creation of pinning sites may be slightly more effective at the lower temperature irradiation. Another possibility is that enhanced defect mobility at the higher temperature irradi-
ation means that point defects (i.e. pinning sites) migrate to grain boundaries faster, leaving less effective pinning sites in the superconducting region. Since both high and low irradiation temperatures lead to a decrease in $\alpha$, this apparently eliminates the possibility that the dependence in irradiation temperature is because of different pinning mechanism/destruction/creation at the different temperatures. Note this is self-consistent with the lack of dependence on irradiation temperature of the crossover temperature for the dominant pinning mechanism.

5.5 Grain boundary-dominated vs. pinning-dominated field regions

With $T_c$ suppression and the creation/destruction of pinning sites eliminated as mechanisms behind the difference in $J_c$ between high and low-temperature irradiation, the two remaining explanations for the higher degradation of $J_c$ in the 423 K irradiated samples are lattice amorphization or grain-boundary widening. Since the highest fluence irradiation performed ($5 \times 10^{16}$ p/cm$^2$) corresponds to a DPA $\approx 0.003$, the creation of a cellular microstructure due to lattice amorphization within grains is not expected [38]. In order to investigate grain boundary disordering, irradiated and control curves of $J_c$ vs. B were analyzed to find the crossover region where grain-boundary limited $J_c$ transitions to pinning-limited $J_c$, as described in Chapter 2. Figure 5-18 shows that at low fluences there is no crossover. As fluence is increased to $1 \times 10^{16}$ p/cm$^2$ and $5 \times 10^{16}$ p/cm$^2$, where noticeable changes in $J_c$ vs. $\theta$ are found, then the crossover appears and increases from $\sim 3$ to $\sim 4$ T between these two fluences. This behavior of increasing crossover field with fluence is consistent with results in the literature [31] and is also observed for the 323 K and 423 K irradiation series in this study.
Figure 5-18: Crossover between grain-boundary limited $J_c$ and pinning limited $J_c$ regimes for 80 K irradiations. Error is represented as the shaded region and are connected between data points to guide the eye to where the crossover occurs. At fluences higher than $5 \times 10^{15}$ p/cm$^2$, a decrease in the irradiated $J_c$ compared to the pristine sample is observed, and the crossover field shifts to higher values of B at higher fluences, reflecting increasing grain boundary disorder becoming dominant over the pinning contributions of the defects at higher field regions.
It should be noted that at the two higher fluences (Figures 5-18 (c) and (d)), irradiation appears to have two distinct effects on the $J_c$ vs. $B$ curves which influence the location of the crossover field. The first effect is a gradual “flattening” of the slope of the curve, which has already been discussed in Section 5.4 as being due to the increase of beneficial pinning centers which lower the value of $\alpha$ and lead to less $J_c$ degradation at higher fields. The second effect is a reduction in $J_c$ over the entire range of applied fields, effectively shifting the irradiated curve downwards. This downward shift represents the effect of grain boundary disorder. As discussed in Chapter 2, as the REBCO sample is irradiated, its grain boundaries act as sinks to defects and become disordered, creating progressively stronger barriers to transport current. As the sample’s grain boundaries become more disordered, the $J_c$ will decrease at all applied fields.

In Figure 5-19, the 80 K and 423 K irradiations at a fluence of $5 \times 10^{16} \text{ p/cm}^2$ are compared. At this fluence, the 423 K irradiation curve has shifted downwards far enough that the crossover field (if it even exists) is beyond the capability of the available testing magnet. The lack of an observed crossover field suggests that $J_c$ over the entire field region shown in Figure 5-19 (b) is grain boundary transport limited. When compared to the low-temperature irradiation at the same fluence with a crossover field of $\sim 5.6$ T, this strongly suggests that grain boundary damage occurs at a much faster rate when a sample is irradiated at elevated temperatures. The large differences in crossover field between cryogenic and heated irradiations at the same fluence indicates that grain boundary widening is likely the most dominant effect behind the globally observed differences in $J_c$ for different irradiation temperatures.
5.6 Effect of annealing

Another observation pointing towards grain boundary widening is the $J_c$ analysis of the annealed cryogenic irradiation sample. While annealing was found to recover part of the critical temperature (as seen in Figure 5-5), annealing was not found to recover critical current, as seen in Figure 5-20. If the decrease in critical current from the ion irradiation had been caused by intragranular damage (such as amorphization), it would be expected that annealing would recover some of this damage as is seen with the critical temperature measurements where Frenkel pairs recombine and the lattice partially recovers. However, in the case of intergranular damage, annealing would simply move more defects towards grain boundaries (which act as particle sinks) and the grain boundary damage would not recover.

Figure 5-20: Angular dependence of annealed vs. un-annealed samples irradiated to $1 \times 10^{16}$ p/cm$^2$ at 80 K.
In radiation materials science, the “gold standard” for measuring and comparing radiation damage has historically been DPA, or the number of displacements per atom. Recent work has challenged this assumption [89], and indicates that while DPA can sometimes be used to roughly predict radiation effects, the irradiation conditions (such as material temperature, dose rate, and radiation uniformity) also play a key role in microscopic damage formation and must be considered. Details of the microscopic damage formation can in turn affect the formation of macroscopic radiation damage features. With this in mind, it is important to note that all radiation studies of commercial REBCO tape use either total neutron fluence or DPA as the measure of damage, with irradiation temperature effects neglected.

In Chapter 5, it was shown that above a threshold proton fluence (somewhere between $1 \times 10^{16}$ p/cm$^2$ and $5 \times 10^{16}$ p/cm$^2$), the temperature at which REBCO samples were irradiated played a critical role in determining the amount of $J_c$ degradation/enhancement of the samples. While high-temperature (423 K) irradiation at a fluence of $5 \times 10^{16}$ p/cm$^2$ led to a significant reduction of $J_c$, cryogenic (80 K) irradiation at the same fluence actually produced an enhancement of the angular minimum $J_c$. While these results would appear to be of great practical importance for the lifetime determination of fusion REBCO electromagnets, a critical question which remains is whether the results are transferable to the conditions in compact, high-field fusion magnet (temperatures ~20 K and neutron irradiation). While ultimately this question must be addressed through dedicated experiments, no facilities currently exist to perform these tests at the current time. Nevertheless, a combination of computational modeling and radiation damage theory will be utilized in this chapter in order to hypothesize about the applicability of the ion irradiation results of this thesis to fusion conditions.
6.1 Assumptions and limitations of the simulations

It is extremely important to note that the modeling and theory work in this chapter have been performed on an “idealized” superconductor. The wide range of (proprietary) tape chemistry and tape manufacturing methods, combined with the multiscale nature of the radiation damage formation processes present a formidable simulation task, far beyond the scope of this thesis. Thus, the purpose of this chapter is not to give quantitative predictions of REBCO lifetime performance in a fusion environment, but rather to compare qualitative trends and determine the whether the temperature-dependent ion-irradiation results of Chapter 5 could be applicable to fusion magnet irradiation conditions. The primary assumptions used in this chapter are as follows:

- In order to simplify the molecular dynamics model (as well as to allow comparison with the sparse literature of REBCO simulation), “pure” (i.e. undoped) YBCO with a stoichiometry of $Y_1Ba_2Cu_3O_7$ was used for all simulations.
- Since oxygen has been shown to be predominantly the most mobile atom in REBCO, all diffusion coefficients have been calculated specifically for oxygen and diffusion effects are assumed to be oxygen dominated.
- “Standard”, isotropic displacement energies were assumed for all atoms in YBCO when determining the primary recoil spectra.

6.2 Simulation workflow

In order to guide the experimental studies in Chapter 3, a simulation workflow was developed by combining DART [118] (a binary collision approximation code), SRIM [102] and MCNP [16] (Monte Carlo codes for ions and neutrons/gammas, respectively), and LAMMPS [119] (a molecular dynamics code), and is illustrated in Figure 6-1: First, the irradiating particle energies are found. For ion irradiation, the HTS superconducting tape geometry and composition is modeled in SRIM, and particles of desired energy and species are sent into the material to determine particle energy at the superconducting layer. For fusion irradiation conditions, the ARC MCNP model described in Chapter 1 was used to determine the neutron energy spectrum at the inner midplane position of the fusion magnet (shown in Figure 1-6). The ion energy or neutron energy spectrum was then passed as an input to the DART code, along with the experimentally measured (for ion irradiation) or predicted (for ARC) fluxes as well as the material composition of YBCO as described above. The DART code would then output a cumulative distribution function of primary knock-on atom (PKA) energies generated by an incident irradiation particle. Using a representative sample of PKA energies generated by DART, molecular dynamics simulations on a $Y_1Ba_2Cu_3O_7$ lattice generated in VESTA were performed using LAMMPS on the Idaho National
Lab Falcon supercomputer\textsuperscript{1}. The results of the LAMMPS simulations were post-processed and analyzed in the OVITO visualization package. Multiple simulations were performed to compare the results of using different ion energies, incident particle directions, and irradiation temperatures with the ultimate goal of understanding the mechanisms behind the experimental results of Chapter 5 and applying them to fusion conditions.

![Simulation workflow](image)

Figure 6-1: Simulation workflow, illustrating inputs and outputs of the analysis codes used in this thesis.

### 6.2.1 Monte Carlo determination of incident irradiation energies

The SRIM package was used to determine the range and stopping power of 1.2 MeV protons from DANTE incident on a 1 \( \mu \text{m} \) Y\(_1\)Ba\(_2\)Cu\(_3\)O\(_7\) layer capped with 2 \( \mu \text{m} \) of silver. As discussed in Section 3.2, the incident protons will slow down ~ 200 keV in the 2 \( \mu \text{m} \) silver cap layer and the average proton energy reaching the superconducting

\textsuperscript{1}This research made use of the resources of the High Performance Computing Center at Idaho National Laboratory, which is supported by the Office of Nuclear Energy of the U.S. Department of Energy and the Nuclear Science User Facilities under Contract No. DE-AC07-05ID14517.

103
layer will be approximately 1 MeV. As seen in Figure 3-11, the PKA energy spectra generated by DART are relatively insensitive to the incident proton energy. This fact, coupled with the relatively small beam energy loss (≈ 100 keV) through the 1 μm YBCO layer, justifies the assumption of a monoenergetic beam in the YBCO layer, greatly simplifying the analysis.

In order to generate the expected neutron energy spectrum at a compact, high-field fusion magnet, the ARC model from Figure 1-5 was evaluated using MCNP on the INL Fission supercomputer. A 23-group energy structure was used to strike a balance between energy resolution, statistics, and computational resources required to run the model. In order to convert the 23-group spectrum into the 100 energy group format which DART requires, the MCNP data was linearly interpolated, as no sharp peaks are expected in the spectrum (see Figure 1-7).

### 6.2.2 Binary collision approximations of PKA energy spectra

When modeling collision cascades from high-energy particles impacting lattice atoms, it is important to distinguish between primary collisions between the incident particle and an atom in the lattice (also known as a “primary knock-on atom” or PKA) and subsequent collisions between displaced lattice atoms. The first type of collisions are dominated by Coulomb scattering (in the case of ion irradiation) or nuclear scattering (in the case of neutron irradiation). While the true ion interaction is governed by a complicated “screened” potential due to the atomic electrons (most often treated with the Lindhard approximation in numerical simulation codes), the “bare” (i.e. no electron) Coulomb scattering cross section given by Rutherford can be used to approximate the cross section. The Rutherford scattering cross section is given as:

\[
\frac{d\sigma}{d\Omega} = \left(\frac{Z_1 Z_2 e^2}{8\pi\epsilon_0 m v_0^2}\right)^2 \text{csc}^4(\theta/2) \tag{6.1}
\]

where \(Z_1\) is the charge of the incident particle, \(Z_2\) is the charge of the target particle, \(m\) and \(v_0\) are the mass and velocity of the incident particle, and \(\theta\) is the scattering angle. The important feature of this equation pertaining to primary collisions is that the cross section is inversely proportional to the square of the incident ion energy. The average PKA energy can be given approximately by [121]:

\[
\bar{T} \approx E_d \ln \left(\frac{\gamma E_i}{E_d}\right) \tag{6.2}
\]

where \(T\) is the recoil energy, \(E_d\) is the energy required to displace a lattice atom (typically ≈ 20 eV), \(E_i\) is the incident particle energy, and \(\gamma\) is a kinematic factor accounting for the mass difference between two elastically scattering particles, given
The result of Equations 6.1 and 6.2 is that as high-energy incident ions pass through a material, they will produce a small amount of PKAs, with (mostly) low recoil energies, and can be modeled using a binary collision approximation in the DART code. Subsequent collisions between the PKA atoms and other lattice atoms will have low impact energies, leading to much more complicated dynamics involving the potentials of all nearby lattice atoms, which must be treated with molecular dynamics codes.

For neutron-ion interactions, elastic nuclear scattering will be the dominant process (with the exception of thermal neutron capture by gadolinium in Gd-doped REBCO, which will be discussed later) leading to displacement of lattice atoms. Compared to Coulomb scattering which is mediated by a long-range force, nuclear scattering collisions can impart a large amount of energy to the recoil atom, with the average PKA energy given by [121]:

\[ F \approx \frac{\gamma E_i}{2} \]  

Simultaneously the neutrons will have an energy spectrum at the coil due to down-scattering, as opposed to the mono-energetic protons. The DART binary collision approximation code was used to determine the PKA energy spectra shown in Figure 6-2 resulting from 1 MeV proton irradiation and neutron irradiation conditions at the ARC toroidal field coil incident on Y1Ba2Cu3O7. The approximately mono-energetic ion irradiation results in a steep-sloped distribution centered at low energies, whereas the neutron irradiation has a much more gradual slope centered at higher energies due to the large spread in the input energy spectrum. The results of Figure 6-2 were used to determine the input conditions to the molecular dynamics model.
Figure 6-2: Comparison of PKA energy cumulative distribution functions resulting from 1 MeV protons and fusion neutrons, generated by the DART code. The large difference in PKA energies for the two types of primary irradiation is due to the different interaction mechanisms governing the collisions (Coulomb scattering vs. nuclear scattering) and the large range of PKA energies from neutron irradiation are due to the wide range of neutron energies incident at the TF coil.

6.2.3 Molecular dynamics simulations

The final step of the simulation was to model the molecular dynamics of the system using LAMMPS (Large-scale Atomic/Molecular Massively Parallel Simulator). LAMMPS takes as input a given atomic structure and calculates the motion of each atom in the lattice due to user-specified pair potentials between the atoms at a given system temperature. LAMMPS was used to simulate oxygen diffusion in the lattice at different temperatures as well as study the effects of defect formation for different PKA particles, energies, and directions at different temperatures.

In order to provide a large enough volume to allow full displacement cascades to propagate, a YBCO unit cell (see Figure 6-3) was constructed using the VESTA visualization software and repeated to create a 40 x 40 x 16 unit cell simulation volume of Y$_1$Ba$_{2}$Cu$_3$O$_7$ (Figure 6-3) with the a, b, and c axes corresponding to the orthogonal [100], [010], and [001] directions. This corresponds to an approximately 15 nm x 15 nm x 19 nm volume of YBCO and was chosen to be large enough to allow cascades up to 10 keV to take place entirely within the volume (see Section 6.3.1) but small enough to allow for a tractable computation time. Periodic boundary conditions were assigned to the faces of the volume.
In order to model the potentials between atoms in the model, the four-part potential of Chaplot [122] was utilized for long-range interactions and the Ziegler-Biersack-Littmark (ZBL) screened potential was used to model short-range (i.e. knock-on) interactions. The Chaplot potential model is given as:

\[
V_{\text{Chap}}(r_{ij}) = \frac{e^2 Z(i)Z(j)}{4\pi \varepsilon_0 r_{ij}} + a\exp\left(\frac{-br_{ij}}{R(i) + R(j)}\right) - \frac{w^6}{r_{ij}} - cD\exp\left(\frac{-n(r_{ij} - r_0)^2}{2cr_{ij}}\right)
\]

(6.5)

where \( r_{ij} \) is the separation distance between the two particles, \( a, b, c, D, n, r_0, \) and \( w \) are coefficients determined through simulation [123], and \( R \) and \( Z \) are the “effective” atomic radii and charges for each atom [124]. The values of the coefficients used are given in Tables 6.1 and 6.2. The first two terms of Equation 6.5 are applied to all pairwise interactions, the third term is applied only to oxygen-oxygen interactions, and the fourth term is only applied to copper-oxygen interactions. The coefficients for \( Z \) and \( R \) were determined assuming perfect \( Y_1Ba_2Cu_3O_{7-\delta} \), i.e. a \( \delta \) value of 0.

<table>
<thead>
<tr>
<th>Coefficient</th>
<th>Value Used</th>
</tr>
</thead>
<tbody>
<tr>
<td>a</td>
<td>1822 eV</td>
</tr>
<tr>
<td>b</td>
<td>12.364</td>
</tr>
<tr>
<td>c</td>
<td>0.5</td>
</tr>
<tr>
<td>D</td>
<td>1.0 eV</td>
</tr>
<tr>
<td>n</td>
<td>8 Å⁻¹</td>
</tr>
<tr>
<td>r₀</td>
<td>1.8 Å</td>
</tr>
<tr>
<td>w</td>
<td>50 eV*Å⁶</td>
</tr>
</tbody>
</table>

Table 6.1: Coefficients used in the Chaplot potential.
Table 6.2: Effective atomic charges and radii used in the Chaplot potential for \( \delta = 0 \).

<table>
<thead>
<tr>
<th>Atom</th>
<th>Z</th>
<th>R (Å)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Y</td>
<td>1.9</td>
<td>1.78</td>
</tr>
<tr>
<td>B</td>
<td>1.5</td>
<td>2.3</td>
</tr>
<tr>
<td>Cu</td>
<td>1.4</td>
<td>1.2</td>
</tr>
<tr>
<td>O</td>
<td>-1.3</td>
<td>1.74</td>
</tr>
</tbody>
</table>

The ZBL potential between two atoms \( i \) and \( j \) at a distance \( r_{ij} \) is represented as:

\[
E_{ij}^{ZBL} = \frac{1}{4\pi\varepsilon_0} \frac{Z_i Z_j e^2}{r_{ij}} \phi(r_{ij}/a) + S(r_{ij})
\]  

(6.6)

where:

\[
a = \frac{0.46850}{Z_i^{0.23} + Z_j^{0.23}}
\]

(6.7)

\[
\phi(x) = 0.18175e^{-3.19980x} + 0.50986e^{-0.94229x} + 0.28022e^{-0.40290x} + 0.02817e^{-0.20162x}
\]

(6.8)

where \( r_{ij} \) is the separation distance between the two particles, \( Z \) is the charge of the particle, and \( S \) is a “switching function” internal to LAMMPS which smoothly transitions the ZBL potential to the longer range potential. The results of the LAMMPS simulations were post-processed using OVITO, an open-source visualization tool [125]. In addition to rendering the positions of each atom in the simulation, OVITO can be used to perform Wigner-Seitz defect analysis as well as measure the size of clusters formed by irradiation.

**Benchmarking the model**

Since the potentials in LAMMPS are up to the user to designate, the model is only as good as the potentials chosen. The potentials specified above are assumed to be valid based on the work of [122] which was in reasonable agreement with experimental results. The model was benchmarked against an oxygen diffusion study [124] performed on YBCO, also using LAMMPS. In order to benchmark the model, the diffusion coefficient at 800 K was calculated through a mean-square-displacement (MSD) analysis as described below in Section 6.4. The calculated diffusion coefficient of \(1.44(±0.05) \times 10^{-8} \text{ cm}^2/\text{s}\) is in close agreement with the value reported in [124] of \(1.1(±0.28) \times 10^{-8} \text{ cm}^2/\text{s}\).
6.3 Defect formation in irradiated YBCO

In order to study defect formation in irradiated YBCO, the molecular dynamics model described above was used to simulate the formation of defect clusters with different initial conditions. The incident primary knock-on atom (PKA) type, PKA energy, PKA direction, and system temperature were all varied. In order to simulate ion irradiation, Ba was chosen as the PKA atom type. The reasoning behind this choice is that the primary mechanism governing the interaction of the beam protons with lattice atoms is Coulomb scattering, with the interaction cross section given in Equation 6.1. The \( Z^2 \) dependence of the cross section means that of the constituents of YBCO, Ba with the highest atomic number of 56 will dominate the interactions with the incident protons.

A PKA direction of [001] (i.e. along the c-axis of the superconductor) was chosen to approximate the orientation of the incident beam normal to the superconducting tape. An observation of Figure 6-2 reveals that the average PKA energy is \( \sim 50-100 \) eV (which is close to the approximation given in Equation 6.2), with a small tail at higher energies. With this in mind, a lower bound for simulation PKA energies was chosen to be 50 eV. An upper bound of 10 keV was chosen to simulate the small fraction of high-energy ion PKAs as well as enable later comparisons with neutron-like conditions, while still allowing a tractable simulation size.

6.3.1 Simulation setup

Before displacing a PKA in the MD model, the simulation volume was relaxed for 100 ps from an initial configuration where the velocity of each atom was randomly selected from a distribution centered at the target simulation temperature. After the relaxation period, an atom at the center of the simulation volume was assigned a velocity along a specified direction corresponding to the PKA energy being studied. The system was then allowed to evolve with a variable timestep, recomputed after every simulation step to allow a maximum of 0.05 Angstrom displacement for any atom in the system. This technique is commonly employed in MD simulations to capture the fast (~fs) dynamics of the ballistic and thermal spike cascade stages, while allowing tractable computation until the cascade thermally quenches on timescales of ~10 ps.

In order to determine the proper MD simulation volume and required simulation time, high-resolution cascades were produced over a wide range of volumes and times to find a system volume which would contain the entire cascade and a simulation time which extended to the end of the thermal quench stage. Figure 6-4 shows the evolution of a 10 keV Ba defect cascade at a temperature of 423 K. The cascade quench phase finishes between 10 and 20 ps, so a standard PKA simulation time of 30 ps was chosen along with the 40 x 40 x 16 unit cell simulation volume.
Figure 6-4: Progression of cascade in YBCO, for Ba [001] PKA of energy 10 keV at a simulation temperature of 423 K. Sites are color coded according to atom type (i.e. Cu is blue, O is red, Ba is green, and Y is gray) and represent either vacancy or interstitials. After approximately 20 ps the defect cluster has fully quenched from the cascade.

After determining the ideal simulation volume and run time, the next step was to determine the "baseline" of interstitial-vacancy Frenkel pairs (FP) produced from the thermal motion of the system (see Figure 6-5). This was required in order to determine the true FP generation from a PKA, as the FP generation output from a PKA simulation includes pairs generated from both the PKA and the thermal motion of the system. To generate the baseline cases, three separate PKA simulations with the incident energy set to 0 eV were run for each temperature being investigated. The mean values of these simulations are shown in Table 6.3. Error was determined by obtaining the standard deviation from the mean.

<table>
<thead>
<tr>
<th>Temperature</th>
<th>Frenkel Pair Production</th>
</tr>
</thead>
<tbody>
<tr>
<td>20 K</td>
<td>0</td>
</tr>
<tr>
<td>80 K</td>
<td>17.7±7.7</td>
</tr>
<tr>
<td>300 K</td>
<td>195±20</td>
</tr>
<tr>
<td>423 K</td>
<td>543±18</td>
</tr>
</tbody>
</table>

Table 6.3: Baseline Frenkel pair production for the entire simulation volume.
6.3.2 Defect formation comparisons

In order to evaluate defect formation for various PKA energies, a Wigner-Seitz defect analysis was performed using the OVITO package at \( t = 30 \) ps using the \( t = 0 \) frame as a reference. Cluster analysis was performed using the baseline FP generation above to determine the cutoff radius for selection of the cluster, effectively “filtering out” the FPs produced by thermal motion from the defects shown below. The simulation results have been re-color-coded to show interstitial sites in red and vacancy sites in blue, although the relative atom sizes have been left the same as the reference model in Figure 6-3. It is important to note that the color-coded sites represent the type of defect (i.e. vacancy or interstitial) at a site, not the actual atom at the defect. For example, a red (i.e. interstitial) Cu site means that the Cu site has an interstitial in it, not that the Cu atom itself is an interstitial.

The first PKA simulations investigated the effect of different PKA energies on defect formation. Figure 6-6 shows a comparison of a 50 eV and 5 keV Ba [001] PKA (starting at the center of the simulation volume). As expected, the 50 eV PKA produces only a few defects, and the much more energetic 5 keV PKA produces a large cluster on the order of \( \sim 5 \) nm. Two interesting features can be observed in the larger cluster. The first is the preferential defect formation along the \( ab \) plane, as evident by the horizontal lines of defects in Figure 6-6 (b). This matches the observation in [124] that especially along the Cu-O chain layers, diffusion occurs much faster along the \( ab \) plane, implying a lower energy barrier for movement along this plane.

The second feature is that a majority of the vacancy sites are oxygen (the small atoms), whereas a majority of the interstitial sites are the larger atoms (mostly Cu1, with some Cu2, Ba, and Y sites), as seen in Figure 6-7. This also matches experi-
Figure 6-6: Comparison of 80 K 50 eV and 5 keV MD simulations. Red particles indicate interstitial sites and blue particles indicate vacancy sites.

Experimental and modeling observations that oxygen is the most mobile species in YBCO [126, 127]. The horizontal rows of interstitial sites observed in Figure 6-7 are Cu1 interstitial sites, signifying the preferential displacement of oxygen along the Cu-O chain layer, and is what gives rise to the “stacked plane” appearance of the defect cluster.

Figure 6-8 compares the extended defects for a 5 keV, [001] directed Ba PKA for simulation temperatures of 80 and 423 K, respectively, to investigate defect formation at the two different irradiation temperatures. Qualitatively, the envelopes of both defect clusters appear to be approximately the same size, although the 423 K cluster has more of a spherical amorphous character, whereas the 80 K PKA has a more “track-like” vertical amorphous region along the c-axis surrounded by the disorder on the Cu-O planes which extends laterally. An observation of even higher energy (10 keV) PKAs (Figure 6-9) confirms this trend.

A slightly more quantitative comparison of the cascade shape can be obtained by comparing the radial distribution functions of both temperature cases. The radial distribution function (represented as g(r)), describes how the density of a structure changes at different radii from a given particle and is the envelope of the histogram created by binning the distance between all pairs in a given volume. Radial distribution functions for both defect clusters in Figure 6-8 were calculated and are shown in Figure 6-10. The presence of sharper peaks in g(r) for the 80 K simulation indicates the existence of a semi-ordered structure, whereas the flatter nature of g(r) for 423 K indicates a more amorphous structure.

A final comparison between the 80 K and 423 K proton irradiation conditions was
performed by computing the number of Frenkel pairs (FPs) generated for a number of different PKA energies (as described in Section 6.3.1). Each energy condition was simulated three times to determine a mean value and standard deviation of FP generation for each energy. Figure 6-11 compares the FP generation at the two temperatures and shows that at low energies, approximately equal numbers of FPs are produced in a cascade, whereas at energies > 1 keV the curves begin to diverge and more FPs are generated at the higher irradiation temperature.

With regards to the proton irradiations, the results shown above indicate that at higher temperatures, the higher energy (E > 1 keV) PKAs produce successively more damage than the low energy PKAs. However, the PKA energy distribution function shown in Figure 6-2 shows that PKA energies above 1 keV are very rare and only make up a few percent of all collisions. Even at the very rare PKA energy of 10 keV shown in Figure 6-11, the ratio between high-temperature and low-temperature FP generation is only ~ 1.5, a ratio which decreases as the PKA energy is lowered. Thus, the effect of irradiation temperature on cluster formation is not expected to play a large role in the $J_c$ degradation effects observed experimentally for ion irradiations.

6.4 Oxygen diffusion in YBCO

Another way in which irradiation could influence the microstructure of YBCO is through radiation-enhanced diffusion of defects to grain boundaries. As a material is irradiated, the simplified radiation-enhanced diffusion coefficient can be given as
Figure 6-8: Comparison of 80 K and 423 K defect clusters produced by a 5 keV, [001] directed Ba PKA in YBCO. Red particles indicate interstitial sites and blue particles indicate vacancy sites.

\[ D_{\text{rad}} = D_v C_v + D_i C_i \]  \hspace{1cm} (6.9)

where \( D_v \) and \( D_i \) are the vacancy and interstitial diffusion coefficients and \( C_v \) and \( C_i \) are the vacancy and interstitial concentration fractions, respectively. As \( C_v \) and \( C_i \) are increased during irradiation, the diffusion coefficient (at a given temperature) is also increased. The results of the previous section indicate that for ion irradiation, defect size is not substantially affected by irradiation temperature, so increases in \( C_v \) and \( C_i \) due to the creation of Frenkel pairs during irradiation would be expected (on short timescales) to be similar for both high and low temperatures. However, the unirradiated diffusion coefficients are highly dependent on irradiation temperature, as will be shown below.

### 6.4.1 Mean-square-displacement simulations

In order to determine the diffusion coefficient when the system is in thermal equilibrium (and is not being irradiated), a mean-square-displacement analysis was performed in LAMMPS. First, the simulation volume was relaxed for 100 ps from an initial configuration where the velocity of each atom is randomly selected from a distribution centered at the target temperature. After the system relaxation, the motion of atoms relative to the reference state is tracked, and the atomic displacement
Figure 6-9: Comparison of 80 K and 423 K 10 keV MD simulations. Red particles indicate interstitial sites and blue particles indicate vacancy sites. The defects at the top of the simulation are result of periodic boundary conditions.

lengths are recorded along each primary direction for each atom and then averaged over all the atoms in the simulation volume to give mean values of displacement in each principal direction (dx, dy, and dz) at each timestep. The total mean-squared displacement (MSD) is determined by adding the squared directional contributions as:

\[
\langle r^2(t) \rangle = \langle dx^2(t) \rangle + \langle dy^2(t) \rangle + \langle dz^2(t) \rangle
\]  

The total MSD can then be plotted vs. time in order to determine the diffusion coefficient. Once the system has reached equilibrium, the MSD should be linear with time, and the diffusion coefficient can be determined from Einstein’s relation [128]:

\[
\langle r^2(t) \rangle = B + 6D\Delta t
\]

where B is a constant, D is the total self-diffusion coefficient, and \( \Delta t \) is the time elapsed. In order to determine statistically significant results, a large total MSD is required, requiring long simulation times, even at high temperatures where the Brownian motion due to thermal vibrations is increased. In order to make the simulations computationally tractable, the simulation volume was reduced to a 10 x 10 x 4 cell (after [124]) and simulations were only possible for temperatures of 700 K and above. Figure 6-12 displays the results of an 800 K MSD simulation to a time of 2500 ps. The first \( \sim 500-1000 \) ps are not in equilibrium, as can be seen from the non-linear slope of the MSD. Thus, the fit to Equation 6.11 was not applied until \( t > 1000 \) ps.
6.4.2 Calculation of diffusion coefficients

Using the method described above, the atomic diffusion coefficients for oxygen (the fastest-diffusing atom in YBCO) were determined for temperatures of 700, 800, 900, and 1000 K. As mentioned above, long computation times make it impossible to directly determine lower temperature diffusion coefficients, but since diffusion coefficients follow an exponential relationship with temperature, the higher-temperature diffusion coefficients can be plotted vs. temperature and fit with a curve used to extrapolate down to the lower temperature diffusion coefficients. Since the (randomly determined) initial conditions of the simulation affect the MSD, three duplicate simulations were performed for each temperature and averaged. Error is represented by the standard deviation of the measurements from the mean. Since diffusion is an Arrhenius process, the diffusion coefficient has a temperature dependence represented as:

\[ D = D_0 \exp \left( \frac{-E_A}{k_B T} \right) \]  (6.12)

where \( E_A \) is the activation energy for diffusion, \( k_B \) is Boltzmann’s constant, and \( D_0 \) is the temperature-independent diffusion constant. It is standard to plot diffusion coefficients on a log plot vs. the inverse of temperature in an “Arrhenius plot” to facilitate a linear fit to the data. An Arrhenius plot for the four simulation temperatures described above is shown in Figure 6-13, along with a fit to the data.

The fit can be used to extrapolate down to temperatures currently inaccessible with molecular dynamics modeling due to the computationally intractable simulation times required. The results of extrapolation down to the irradiation temperatures used in the experiments allow for a comprehensive understanding of the diffusion process at various conditions.
this thesis are presented in Table 6.4 and show an enormous (27 order of magnitude) decrease in the diffusion coefficient value between the heated and cryogenic irradiations.

<table>
<thead>
<tr>
<th>Temperature</th>
<th>Diffusion Coefficient D(T)</th>
</tr>
</thead>
<tbody>
<tr>
<td>80 K</td>
<td>$3.1 \times 10^{-38}$ cm$^2$/s</td>
</tr>
<tr>
<td>423 K</td>
<td>$1.6 \times 10^{-11}$ cm$^2$/s</td>
</tr>
</tbody>
</table>

Table 6.4: Diffusion Coefficient Extrapolations

It is worth re-iterating that the results in Table 6.4 are extrapolations which are themselves based on simulations of an ideal material with several approximations. Thus, the absolute values presented below are very rough approximations of the true oxygen diffusion coefficient in the REBCO which was irradiated in Chapter 5. However, the large relative difference between the cryogenic and heated irradiations points to greatly enhanced radiation-assisted diffusion at the higher temperature, which is consistent with the hypothesis that enhanced grain boundary disordering occurs at higher temperature irradiations due to increased diffusion of defects to the grain boundaries which act as sinks to the defects. Over a given time period $t$, the distance a particle will diffuse ($d$) can be approximately given as:

$$d \approx \sqrt{Dt}$$

The high fluence ($5 \times 10^{16}$ p/cm$^2$) irradiations took approximately 80 minutes (4800 s). Using this time, the approximate average diffusion distances for the 80 K and 423
K irradiations can be calculated. For 80 K, $d = 1.2 \times 10^{-9}$ A, meaning that there is essentially no diffusion at all. However, at 423 K, $d = 2.8$ μm, which is on the order of the grain size in modern REBCO conductors [129]. While these numbers are approximations, they illustrate the extreme differences between diffusion at the two different irradiation temperatures. In fact, based on the experimental results obtained on the annealed sample (see Figure 5-20), it would be expected that the true diffusion coefficient at 423 K is lower, as the sample annealed at 423 K after cryogenic irradiation retains a higher $J_c$ than the sample irradiated to the same fluence while being held at 423 K during irradiation. The lack of grain boundary widening after annealing is highly encouraging, as it indicates that fusion magnets brought up to room temperature for maintenance will not suffer $J_c$ degradation and will retain the benefits of cryogenic irradiation.

The results of this section and the previous section analyzing Frenkel pair generation both support the experimental evidence for grain-boundary disorder as the dominant mechanism limiting $J_c$ transport for REBCO irradiated with ions at high temperatures. Since the diffusion coefficient is already essentially zero at 80 K, it would not be expected that irradiation at 20 K would have substantially different behavior.

### 6.5 Applicability of REBCO ion irradiation results to fusion neutron irradiation

The above results from both simulation and experiment suggest that cryogenic irradiation of REBCO at fusion conditions (low temperature and high field) could substantially improve the survivability of REBCO in an irradiation environment. The
obvious question following from these results is whether they hold for neutron irradiation as well as ion irradiation. Unfortunately no cryogenic neutron irradiation facilities currently exist to test this. While ultimately a facility will need to be built to perform these tests on actual REBCO samples, the following section will use the molecular dynamics model created above to model neutron-like conditions and make predictions about the applicability of the ion irradiation results to neutron irradiation.

As illustrated in Figure 6-2, a substantial difference between neutron and ion irradiation is the difference in PKA energy spectra for the two types of irradiation, which is due to the aforementioned different interaction mechanisms between ions and neutrons with the lattice atoms. In addition to having substantially higher-energy PKA atoms, radiation by a fusion neutron spectrum also produces a PKA spectrum with a much flatter slope than for ion irradiation, meaning that there is a wide range of PKA energies which could be generated. Another difference (again due to the interaction mechanism) is that the nuclear scattering cross section is not dictated by the atomic number as with the cross section for ion interactions and is instead sensitive to the particular isotope. A third important difference is that unlike ion-atom interactions which are all governed by some form of Coulomb scattering, there are a number of different nuclear scattering mechanisms besides elastic scattering, such as inelastic scattering, neutron multiplication, and neutron capture reactions. A full simulation of these effects is beyond the scope of this thesis and in any case would be pointless on the idealized YBCO model described above. Since neutron reaction cross sections are highly dependent on the isotopic composition of the exact material being studied full simulations would only be valid if the exact composition of the commercial REBCO tapes was known. For the following analysis, the only neutron-atom inter-

$$D = D_0 \exp \left( \frac{-E_A}{k_B T} \right)$$

$$D_0 = (2.65 \pm 0.86) \times 10^{-5} \text{ [cm}^2/\text{s]}$$

$$E_A = 0.52 \pm 0.03 \text{ [eV]}$$
action mechanism considered will be elastic scattering, except for a brief discussion of thermal capture to gadolinium discussed at the end of this chapter.

6.5.1 Defect formation

An observation of the neutron energy spectrum at the ARC magnet (shown in Figure 1-7) reveals that a majority of the neutrons reaching the toroidal field magnet have energies between $\sim 100$ keV and 10 MeV, with a broad peak around a few MeV. An observation of neutron elastic scattering cross sections from the ENDF/B VII libraries [130] for the principal isotopes of elemental YBCO reveals that in this energy range, the scattering cross sections (with the effects of resonance regions averaged out) are approximately within a factor of 2 of each other ($\sim 10$ b). Thus, the lattice atom chosen for the PKA was chosen to be oxygen due to its predominance in the stoichiometry of $\text{Y}_1\text{Ba}_2\text{Cu}_3\text{O}_7$.

Since neutron interactions produce a much more isotropic distribution of PKA directions, a comparison was performed on oxygen PKAs directed along the three major crystallographic directions, shown in Figure 6-14. Although there is possibly a slight reduction in FP production for the [010] (i.e. b-axis) directed PKA at the high energy, the defect production is within overlapping error bars. Thus, for the purposes of this study, the [001] direction will be used for the remaining oxygen PKA simulations.

![Figure 6-14: Frenkel pair production (per PKA) vs. PKA energy for O [001], [010], and [100] directed PKAs at 20K.](image)

Figure 6-14 shows a comparison of Frenkel pair production per PKA for Ba and O [001] PKAs over the lower range of energies applicable for neutron irradiation, at the fusion-relevant irradiation temperature of 20 K. At low energies, the FP production appears
to be similar, while at high energies FP production from Ba PKAs starts to slightly outpace production from O PKAs. If this trend continues at even higher energies, it would indicate that neutron irradiations would produce less average Frenkel pairs per collision. From a $J_c$ degradation standpoint, this would be favorable, as smaller defect clusters would act as more efficient pinning sites at high fields, and less total concentration of Frenkel pairs would lead to less radiation-enhanced diffusion to grain boundaries.

A final comparison is shown of Frenkel pair production per PKA vs. PKA energy for simulation temperatures of 20 K, 300 K, and 423 K. The purpose of the two higher temperatures is that all $J_c$ studies with neutron irradiation on commercial REBCO tapes have been performed at temperatures between 300 and 423 K. As shown in Figure 6-16, there appears to be a slight trend towards more Frenkel pair production at lower temperature, but the high-energy results at all three temperatures are within the error bars of each other, so Frenkel pair formation would not appear to be a large mechanism driving different $J_c$ degradation rates at different irradiation temperatures.

As with the ion irradiations, it would appear that the primary mechanism which could be responsible for differences between cryogenic and high-temperature irradiations would be large differences in the thermal diffusion coefficient leading to highly divergent rates of radiation-enhanced diffusion. Using the fit to the diffusion curve in Figure 6-13, the extrapolated diffusion coefficients for YBCO at 20 K and 300 K are shown in Table 6.5 (along with the previously calculated diffusion coefficient for 423 K for comparison).
Figure 6-16: Frenkel pair production per PKA vs. PKA energy for O [001] PKAs at 20K, 300K, and 423K.

<table>
<thead>
<tr>
<th>Temperature</th>
<th>Diffusion Coefficient D(T)</th>
</tr>
</thead>
<tbody>
<tr>
<td>20 K</td>
<td>$5.9 \times 10^{-13}$ cm$^2$/s</td>
</tr>
<tr>
<td>300 K</td>
<td>$4.4 \times 10^{-14}$ cm$^2$/s</td>
</tr>
<tr>
<td>423 K</td>
<td>$1.6 \times 10^{-11}$ cm$^2$/s</td>
</tr>
</tbody>
</table>

Table 6.5: Diffusion coefficient extrapolations to temperatures relevant to neutron irradiation studies as well as fusion conditions.

The diffusion coefficient at 20 K is effectively zero, and the diffusion coefficient at room temperature is almost three orders of magnitude below the diffusion coefficient at 423 K. This suggests that radiation-enhanced diffusion will be extremely sensitive to sample temperature during irradiation—a fact that all studies to date of neutron irradiated REBCO have neglected to address. While there could be other mechanisms causing different rates of $J_c$ degradation between ion and neutron-irradiated REBCO, the above results indicate that the same mechanisms behind the experimentally observed importance of sample temperature control for ion irradiations also apply to neutron irradiations.

### 6.5.2 Gadolinium effects

Although not modeled in the above simulations, a topic of practical importance for fusion magnets is the effect of thermal neutron irradiation on gadolinium. Modern REBCO coated conductors are often doped with Gd to increase critical current performance [131]. Unfortunately, the isotopes of Gd-155 and Gd-157 have two of
the highest thermal (and epithermal) neutron capture cross sections of $10^4$ to $10^5$ b, compared to the $\sim 10$ b cross sections for elastic scattering of the other component isotopes of REBCO. After the Gd nucleus absorbs the neutron, it is left in an excited state and will emit a gamma particle to return to the ground state. The average recoil energy to the Gd nucleus from the gamma particle is on the order of 20-40 eV, enough to sometimes displace the Gd atom from its lattice site and create point defects [33]. This leads to a situation where the true PKA spectrum of neutron-irradiated REBCO could be weighted more heavily towards lower energy recoils (of specifically Gd) and has been raised as a concern for radiation damage in neutron environments by several studies [33, 114, 132]. However, all irradiation studies of Gd-REBCO to date have been performed at elevated temperatures, typically between 323 K and 353 K. Based on the results shown in this thesis, the creation of large amounts of isolated Frenkel pair defects would lead to high amounts of grain boundary damage at these high irradiation temperatures. At low temperature irradiation though, the defects would be immobile and would not contribute to grain boundary disordering. Thus, it would be a very interesting experiment for future work to compare high and low-temperature Gd-REBCO irradiations with neutrons. If it does turn out to be the case that low-temperature irradiation obviates the problems described in [33, 114, 132], this would be highly advantageous for fusion magnets, as the higher-performance Gd-REBCO could be used to construct fusion magnets.
Chapter 7

Conclusions and Recommendations for Future Work

Results for the $J_c$ changes of proton-irradiated REBCO tapes at different fluences and irradiation temperatures have been presented in this thesis. Ion irradiation at cryogenic temperatures was found to substantially reduce the amount of $J_c$ degradation in the REBCO samples irradiated to high fluences. This result is of great importance to superconducting REBCO magnets in fusion applications where the radiation will occur at $T < 80$ K. An analysis of temperature, field, and angle dependencies of $J_c$ suggests that the microstructural mechanism behind the differences of $J_c$ with irradiation temperature for a given fluence is two-fold. The first effect is that the creation of effective weak, uncorrelated pinning sites appears to be slightly enhanced at the lower temperature irradiations. This effect is most likely caused by the decreased defect mobility at low irradiation temperatures, leading to the preferential creation of point defects or small defect clusters which act as more effective weak pinning sites. The practical result of this effect is that REBCO performance at high fields is increased, partially canceling out the detrimental effects due to lattice disorder from the irradiation. The second, larger, effect is that higher-temperature irradiations cause significantly more grain boundary damage, evidenced by the large decrease in $J_c$ over all fields for the high temperature irradiations. This effect can be explained by a much larger oxygen diffusion coefficient at high temperatures (obtained through molecular dynamics simulations), leading to the enhanced migration of defects to grain boundary sinks during irradiation.

Molecular dynamics simulations suggest that the same mechanism (i.e. enhanced grain boundary disorder) behind the experimentally observed importance of sample temperature control for ion irradiations also applies to neutron irradiations. This motivates a re-evaluation of previous REBCO neutron irradiation studies at temperatures between 323 K and 353 K, specifically with regards to predictions about REBCO lifetimes in a fusion environment. The work in this thesis suggests that at cryogenic temperatures, the $J_c$ degradation observed in these studies could be sub-
stantially ameliorated, leading to longer estimates for tape lifetime. Based on this conclusion, the author strongly recommends the construction of a cryogenic neutron irradiation facility for the purpose of experimentally extending the results of this work to neutron irradiation studies.

7.1 Major accomplishments of this thesis

- The neutronics of a compact, high-field tokamak reactor (ARC) were assessed to determine the effect of neutron irradiation on the overall design of compact fusion reactors.

- A 1 MV tandem accelerator was refurbished and upgraded in order to enable the irradiation of REBCO samples with light ions.

- A temperature-controlled target stage was constructed to allow for both cryogenic and heated irradiations of REBCO samples, along with a precision beam imaging system to ensure uniform irradiation of samples.

- Comprehensive scans of critical current vs. field, temperature, and field angle were performed on irradiated and unirradiated REBCO samples to assess the effect of different radiation fluences and temperatures on sample degradation.

- Evidence was presented to show the importance of irradiation temperature when determining REBCO lifetimes.

- Molecular dynamics simulations of YBCO were carried out to investigate the microstructural mechanisms responsible for the experimentally observed temperature-dependent irradiation effects.

7.2 Implications for future devices

From the ARC reactor study, it is clear that the operational lifetime of the TF coils will play a key role in determining the economics of compact, high-field fusion reactors. Unfortunately, since neutron damage has the most detrimental effect at the high-field region in the inner midplane, any attempts to reduce the amount of radiation arriving at the magnet by simply adding more shielding will have a ripple effect throughout the entire device. Adding more shielding to the inner midplane region will push the major radius of the plasma farther from the magnet itself. Due to the magnetic field dropoff with radius from the toroidal field coil, this requires a larger TF coil to provide the same on-axis field to the plasma, requiring more inner leg space. The increase in major radius will increase fusion power, which is beneficial from a global sense but also increases the neutron flux to the magnet, requiring more shielding to have the same effect. In summary, one cannot simply add more shielding
to the inner leg without a substantial increase in device size, and thus cost. Since one of the key drivers behind designing compact, high-field fusion reactors is capital cost reduction of the fusion island, all attempts must be made to limit the amount of shielding required at the inner leg of the tokamak.

With this in mind, it becomes imperative to know to a high degree of certainty the lifetime of the REBCO in the TF coils. Unlike REBCO irradiation studies for large tokamak designs such as ITER or DEMO where there is ample room for shielding, REBCO irradiation studies for compact, high-field tokamaks must be undertaken to determine the absolute maximum tolerable fluences to REBCO under fusion-relevant conditions to allow for the smallest (and thus cheapest) device possible. The results of this thesis suggest that cryogenically irradiated REBCO will have higher tolerance to radiation than REBCO irradiated at room or elevated temperatures, implying that the current “standard” high-energy radiation limit of $3 \times 10^{18}$ n/cm$^2$ to REBCO is too conservative. Since REBCO lifetime is a key factor determining the economics of compact fusion reactors, it is worth determining the “true” lifetime of REBCO in fusion conditions. In mature industries such as natural gas turbine generators, improvements of even a few percent to turbine efficiency are sought out [133] and justified by the large economic advantage that they would confer to the industry. It is not inconceivable that in a future fusion energy economy REBCO lifetime will have a similar importance.

A key result of this study is that cryogenic neutron irradiations are required to assess the true performance of REBCO coated conductors for use in a compact, high-field fusion reactor. While a limited number of neutron irradiation studies have been performed on modern coated conductors, all of these studies have been performed at elevated temperatures, and subsequent analysis of the irradiated tapes has been performed at higher operating temperatures and lower operating fields than will be present in a compact fusion reactor. As the lifetime of REBCO coated conductors in a fusion environment will have a large influence on power plant design, it is critical to accurately determine this parameter.

7.3 Suggestions for future work

While the work in this thesis has established the importance of sample temperature control during REBCO irradiation, there is still much work remaining to carry the work to its practical conclusion of determining (and possibly extending) the lifetime of REBCO used in fusion magnets. The following section outlines a few suggestions for the most “mission critical” work pertaining to this goal.
7.3.1 Cryogenic, in-core reactor irradiation

By far the most important follow-on work to this study would be in-core fission reactor irradiations of commercial REBCO conductors under cryogenic conditions. While cryogenic neutron irradiations were performed on Nb$_3$Sn during the ITER conductor qualification process, the facilities used have all been shut down. Recently, a cryogenic test loop has been operated at the OSU Research Reactor [134], but at the time of this thesis the test loop was not operational. The MIT Reactor Lab used to run an in-core liquid-nitrogen-cooled loop in its 6 MW research fission reactor and is another good option for the construction of a cryogenic REBCO neutron irradiation facility.

7.3.2 High-fidelity analysis of neutron-irradiated samples

As a corollary to the need for cryogenic neutron irradiations, the results of this thesis have shown that it will also be necessary to obtain high-fidelity critical current measurements over a wide field, temperature, and field angle parameter space. Unfortunately, since neutron-irradiated tapes become significantly activated, they cannot be externally transported and analyzed in the RRI SuperCurrent machine due to contamination concerns. In the future, it would be highly beneficial to have a dedicated SuperCurrent measurement system which could be operated in a radiation environment in order to obtain high-fidelity measurements of neutron-irradiated REBCO.

7.3.3 In-situ accelerator $I_c$ analysis

An important question unanswered by this thesis is whether simply warming cryogenically-irradiated REBCO samples to room temperature is enough to partially anneal the sample and change the pinning structure inside the irradiated tape. Since fusion magnets will be operated and irradiated concurrently at cryogenic temperatures without warming at all, it is important to determine whether any such annealing could have played a role in the results of this thesis. In the same way that this thesis used accelerator irradiations to quickly and cost-effectively determine whether irradiation temperature would be important in neutron irradiations, it would be feasible to install an in-situ transport measurement system at the end of the DANTE beamline to investigate the effects of in-situ irradiation damage at cryogenic temperatures.

7.3.4 Further annealing studies

It has been postulated by some studies [135, 136] that the annealing of irradiated REBCO can return the superconductor at least partially to its unirradiated state. While the results of the single annealing study performed for this thesis indicates
that annealing is much more effective on restoring $T_c$ than $J_c$, the possibility of “re-generating” radiation-damaged TF coils motivates more detailed study into different types of annealing. An interesting possibility would be annealing at high temperature (i.e. 500-900 K) in an oxygen-rich environment to attempt to anneal out the grain boundary disorder without causing de-oxygenation of the REBCO crystal.

7.3.5 TEM comparisons of ion and neutron irradiated tapes

A body of work was carried out in the early 1990’s on TEM characterization of ion and neutron irradiated YBCO single crystals [79]. While this work was useful in developing an intuition for intragrain radiation damage, it is largely inapplicable to today’s modern coated conductors with a wide variety of tape chemistries and artificial pinning center inclusions. More importantly, modern coated conductors are composed of many, carefully aligned grains and (as discussed in this thesis) intergrain transport is of critical importance in REBCO irradiation studies. With this in mind, it would be advantageous to repeat the same type of studies reported in [79], but with modern REBCO conductors which have been irradiated in cryogenic and heated conditions.

7.3.6 Isolating thermal neutron effects on different tape chemistries

A final area of important future research is a systematic investigation of the effects of thermal neutron capture in gadolinium. In the past few years, many tape manufacturers have started doping YBCO tapes with gadolinium to increase tape performance. While thermal neutrons do not possess enough energy to displace atoms themselves, Gd-155 and Gd-157 have extremely high thermal neutron capture cross sections, and when the excited Gd nucleus decays back to the ground state, the nucleus can recoil at energies high enough to displace the Gd atom from the lattice, creating a point defect. Since exact REBCO composition is often proprietary and not released by manufacturers, it would be advantageous to obtain custom-made REBCO tapes with different fractions of Gd doping to investigate the severity of radiation effects due to thermal neutron interactions with Gd, at both high and low irradiation temperatures. Since Gd doping is becoming more common to increase tape performance, it will be critical to answer the question of whether REBCO used to fabricate fusion magnets can be allowed to tolerate Gd doping, and if so what fraction of Gd doping would be acceptable.
Appendix A

Detailed DANTE Repair Information

A.1 Inductor design

During the course of accelerator repair, it was necessary to replace the ~1 H matching inductor at the top of the voltage multiplier circuit. Due to its large inductance value and unique size/voltage requirements, the inductor had to be fabricated. First, a formula from "Handbook of electronics calculations for engineers and technicians" [137] was used to calculate the dimensions and turns required, as shown in Figure A-1.

\[
L = \frac{(0.315)r^2N^2}{6r + 9l + 10d}
\]

\( L \) = inductance, in uH
\( N \) = total # turns
\( r, l, d \) = all length values in cm

**For \( l \), it was assumed that because the "dividers" in the segmented design are thin enough, the 23 segments in series are coupled and can effectively be treated as one segment with total turns \( N \) and

\[
l = 23l_{seg} + 22l_{div}
\]

Figure A-1: Calculation of required dimensions and turns for replacement 1 H inductor in DANTE.
A 23-segment inductor former was drafted using SolidEdge (see Figure A-2), and was fabricated from Delrin, an electrically insulating material.

Figure A-2: CAD drawing of the former used to wind the inductor.

Approximately 4,000 ft of 27 gauge, NEMA MW16-C magnet wire insulated with heavy polyimide was used to wind the 7,820 total turns (340 turns/segment) required for the inductor. The final calculated inductance as $\sim 0.87 \, \text{H}$. The fabricated inductor is shown installed in the DANTE terminal in Figure A-3.

A.2 Normal operating conditions

After repairing the ion source, normal operating conditions for the accelerator slightly changed compared to the values presented in the previous work of Blackburn [90]. In order to provide a point of reference for future users of DANTE, typical operation values are presented below in Table A.1:

Figures A-4 and A-5 are photographs of the DANTE control system and magnet supplies taken during the March 29, 2017 irradiations that the values in Table A.1 were taken from. During operation, it was found that the source provided a steadier beam current when the filament, arc, and plasma electrode supplies were run in voltage control mode. It is important to note that although the control program does not display a status bar for plasma electrode voltage, the voltage control does work (as verified by the physical indicator on the supply). Operationally, it was found that the mass flow controller would "stick" at low settings, and would sometimes take several seconds to start puffing gas into the source.
Table A.1: List of significant DANTE operational parameters, recorded during March 29, 2017 irradiations. Nominal beam current on target was ~300 nA and could be increased/decreased by adjusting the filament current. Note that values quoted are the actual values (represented in green text in Figure A-4), not the set point values.
Figure A-3: Final installation of custom-built inductor.

Figure A-6 displays a typical oscilloscope trace from the terminal switching power supply. The shape and behavior of this trace provide a metric of how well the terminal is performing. The scope trace should be approximately a square wave and should be stable to ensure reliable and safe terminal operation. A "jagged" or unstable scope trace indicates the possible presence of arcing in the acceleration tank/column and means that the terminal should be turned off immediately.

Figure A-4: DANTE control screen.
Figure A-5: Magnet power supplies providing current to the selection (top), horizontal (second down) and vertical (third down) steering magnets, and focusing quadrupoles (bottom two supplies).

Figure A-6: Typical trace of terminal switching power supply signal.
Appendix B

Molecular Dynamics Simulations

B.1 LAMMPS input files

The input files used to generate the molecular dynamics results in Chapter 6 are presented below. Since simulation volumes were large, a Python script was used to generate the repeating unit cell structure of YBCO which was saved to the "*.dat" files used in the LAMMPS script to define the atoms in the volume. Since the fourth term in the Chaplot potential (Eq. 6.5) was not pre-defined in LAMMPS, another Python script was used to generate a table of force/potential values manually for the pairwise (3,5), (4,5), and (5,5) interactions.

B.1.1 MSD input file

```
# -------------------------------- PKA simulation for YBCO --------------------------------

variable myT equal 800

# ---------------------------------- INITIALIZATION ----------------------------------

units metal
boundary p p p
dimension 3
atom_style charge
atom_modify map hash
read_data YBCO_10_10_4.dat
region inter block -17.000000 17.000000&
                  -17.100000 17.100000&
                  -21.380000 21.380000 units box

region outeratom region inter
region outeratom subtract all interatom
```
compute interT interatom temp
compute outT outeratom temp

# ------------------ Force Field ------------------

pair_style hybrid/overlay buck/coul/long 9.990001 9.990001
     zbl 0.9 1.8 table linear 1000
pair_coeff *  * buck/coul/long 1e-30 1e-30 0.0
pair_coeff 1 1 buck/coul/long 1822.000000 0.287933 0.0
pair_coeff 1 2 buck/coul/long 1822.000000 0.329990 0.0
pair_coeff 1 3 buck/coul/long 1822.000000 0.241022 0.0
pair_coeff 1 4 buck/coul/long 1822.000000 0.241022 0.0
pair_coeff 1 5 buck/coul/long 1822.000000 0.284698 0.0
pair_coeff 2 2 buck/coul/long 1822.000000 0.372048 0.0
pair_coeff 2 3 buck/coul/long 1822.000000 0.283080 0.0
pair_coeff 2 4 buck/coul/long 1822.000000 0.283080 0.0
pair_coeff 2 5 buck/coul/long 1822.000000 0.326755 0.0
pair_coeff 3 3 buck/coul/long 1822.000000 0.194112 0.0
pair_coeff 3 4 buck/coul/long 1822.000000 0.194112 0.0
pair_coeff 3 5 buck/coul/long 1822.000000 0.237787 0.0
pair_coeff 4 4 buck/coul/long 1822.000000 0.194112 0.0
pair_coeff 4 5 buck/coul/long 1822.000000 0.237787 0.0
pair_coeff 5 5 buck/coul/long 1822.000000 0.281462 0.0
pair_coeff 1 1 zbl 39.000000 39.000000
pair_coeff 1 2 zbl 39.000000 56.000000
pair_coeff 1 3 zbl 39.000000 29.000000
pair_coeff 1 4 zbl 39.000000 29.000000
pair_coeff 1 5 zbl 39.000000 8.000000
pair_coeff 2 2 zbl 56.000000 56.000000
pair_coeff 2 3 zbl 56.000000 29.000000
pair_coeff 2 4 zbl 56.000000 29.000000
pair_coeff 2 5 zbl 56.000000 8.000000
pair_coeff 3 3 zbl 29.000000 29.000000
pair_coeff 3 4 zbl 29.000000 29.000000
pair_coeff 3 5 zbl 29.000000 8.000000
pair_coeff 4 4 zbl 29.000000 29.000000
pair_coeff 4 5 zbl 29.000000 8.000000
pair_coeff 5 5 zbl 8.000000 8.000000
pair_coeff 3 5 table Potential_35.table LS_YBCO 9.990001
pair_coeff 4 5 table Potential_45.table LS_YBCO 9.990001
pair_coeff 5 5 table Potential_55.table LS_YBCO 9.990001
kspace_style pppm 0.0001
neigh_modify delay 0 every 1 check yes

group oxygen type 5

###------------------------Plot energy/force vs. distance r------------------------
thermo 100
thermo_style custom step pe ke press lx lylz temp pxx pyypzz
minimize 1e-10 1e-10 5000 10000

###------- Relax. at Target Temp for 100 ps ---------###
reset_timestep 0
timestep 0.001
velocity all create ${myT} 8200 mom yes
fix 1 all nvt temp ${myT} ${myT} 0.1
run 100000

#---------------- MSD -------------------------------------#
compute Omsd oxygen msd com yes
variable OMSDX equal "c_0msd[1]"
variable OMSDY equal "c_0msd[2]"
variable OMSDZ equal "c_0msd[3]"
variable OMSDAAll equal "c_0msd[4]"
thermo 100
thermo_style custom step pe ke press lx lylz temp &
fix 2 all print 1000&
"${OMSDX} ${OMSDY} ${OMSDZ} ${OMSDAll}" &
file MSD.data screen no
run 3000000

B.1.2 PKA input file

#---------------- PKA simulation for YBCO ------------------#
variable myT equal 80

#---------------- INITIALIZATION --------------------------#
units metal
boundary p p p
dimension 3
atom_style charge
atom_modify map hash
read_data YBCO_40_40_16.dat

#------- select PKA atom, define boundary region ---------#
variable PKAEnergy equal 10000 #units eV
variable PKAID equal 2 # ID of 2 is Ba atom
group PKAAtom id ${PKAID}
variable PKAmass equal mass[v_PKAID]
variable dispx equal -x[${PKAID}]
variable dispy equal -y[${PKAID}]
variable dispz equal -z[${PKAID}]
displace_atoms all move ${dispx} ${dispy} ${dispz} units box
variable PKAVel equal &
   "sqrt(v_PKAEnergy*19296.823743/v_PKAmass)"
variable pkaVx equal 0
variable pkaVy equal 0
variable pkaVz equal "-v_PKAVel"
region inter block
   -72.000000 72.000000
   -72.400000 72.400000
   -89.520000 89.520000 units box
group interatom region inter
group outeratom subtract all interatom
compute interT interatom temp
compute outT outeratom temp

# --------------- Force Field --------------------------------

<same as MSD input file>

#-------------------Plot energy/force vs. distance r ----------------

<same as MSD input file>

#------------------- Relax. at Target Temp for 100 ps ----------------
reset_timestep 0
timestep 0.001
velocity all create ${myT} 8636 mom yes
fix 1 all nvt temp ${myT} ${myT} 0.1
run 100000
unfix 1

#------------------- PKA ensemble -------------------
fix 1 all nve
fix 2 outeratom temp/rescale 1 ${myT} ${myT} 2 1
fix 3 all dt/reset 1 1.0e-6 1.0e-3 0.05 units box

# Assign velocity to PKA atom
velocity PKAatom set ${pkaVx} ${pkaVy} ${pkaVz} units box

# Compute pka atom energy and all atom energies
compute pkaKinetic PKAatom ke/atom
compute KineticAll all ke/atom
dump 1 all custom 10000 cascade.data.* id&
    type x y z c_KineticAll
# Run simulation for 30 ps to let cascade settle down
run 30000
References


[41] HEIKE Kamerlingh Onnes. Investigations into the properties of substances at low temperatures, which have led, amongst other things, to the preparation of liquid helium. *Nobel lecture*, 4, 1913.


