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The Impact of Superconducting Properties of Micron-Scale Masked Proton Irradiation on $BaTiO_3$-Doped YBCO Film

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Abstract: This study investigates the effects of 60 keV proton irradiation on $BaTiO_3$-doped $YBa_2Cu_3O_{7-\delta}$ (YBCO) films using masks with micron-scale holes to create controlled defect patterns aimed at enhancing superconducting properties. Contrary to expectations, masked irradiation resulted in a reduction in the critical current density (J_c), while unmasked irradiation demonstrated improvement, consistent with previous studies. Notably, no improvement was observed at 2 T around liquid nitrogen temperature. These observations highlight the challenges of employing micron-scale masks in defect engineering and underscore the need for further refinement to achieve the desired performance enhancement. Insights from this study contribute to advancing defect engineering techniques for improving YBCO's performance in high-field applications, including fusion energy systems.

Keywords: YBCO; proton irradiation; fusion

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1. Introduction

Second-generation coated conductors (CCs) based on biaxially textured $REBa_2Cu_3O_{7-\delta}$ (REBCO, where RE represents rare-earth elements such as Y, Gd, Sm, or others) films have garnered significant attention due to their exceptional properties, particularly their high T_c and robust in-field J_c. These features make REBCO films invaluable for advanced magnetic applications, including those requiring the generation of intense and stable magnetic fields [1]. Among these materials, YBCO was the first material discovered to exhibit superconductivity above the boiling point of liquid nitrogen [2], and YBCO tapes have emerged as a cornerstone for fusion energy technologies. Fusion reactors, such as tokamaks, rely on high-performance high-temperature superconductors to generate and sustain the powerful magnetic fields needed for plasma confinement. The ability of YBCO tapes to carry high current densities without resistance while operating under liquid nitrogen cooling conditions highlights their potential as indispensable materials for fusion energy systems [3].

Despite their high production costs, YBCO tapes are particularly appealing for use in fusion magnets due to their unparalleled combination of superconducting properties and temperature stability. In response to cost challenges, significant advancements have been made in production techniques, most notably through the adoption of the trifluoroacetate–metal organic deposition (TFA-MOD) process in 2005. This process has facilitated higher production rates and substantially reduced costs, leading to the commercial availability of YBCO tapes at more competitive prices [4]. However, a critical challenge remains: enhancing the in-field J_c of these tapes to meet the stringent requirements of fusion magnets. In tokamak reactors, high-current conductors in the kiloampere (kA) range are essential

to limit voltage spikes during rapid discharge events [5]. Thus, developing methods to optimize the in-field J_c of YBCO tapes has become a vital area of research, with the goal of ensuring their viability for use in next-generation fusion energy systems.

Another significant challenge lies in the irreversibility field (H_{irr}). For instance, J_c decreases substantially under high magnetic fields [6]. This limitation calls for further advancements in superconducting properties to meet the rigorous demands of fusion applications. Enhancing the irreversibility field (H_{irr}) while maintaining high J_c in this regime is crucial for enabling practical applications. The introduction of controlled defects has been shown to improve both J_c and irreversibility field (H_{irr}). Recent progress in YBCO with nano-columnar defects has demonstrated an increased irreversibility field (H_{irr}) reaching approximately 10–11 T at 77 K [7]. Furthermore, melt-textured REBaCuO with nano-particles at elevated temperatures, such as liquid oxygen temperature (90.2 K), has exhibited consistently high J_c [8].

One promising approach to enhance J_c involves the introduction of artificial pinning centers (APCs). APCs have been shown to effectively reduce vortex motion, a major contributor to resistance in superconducting materials under high magnetic fields [9]. Techniques for introducing APCs typically involve particle irradiation, which can be applied to various superconducting materials, including YBCO. For instance, neutron and ion irradiation at different energy levels have been widely employed to create flux pinning sites, providing valuable insights into superconducting ground states and enhancing the performance of YBCO films [10,11]. Among these techniques, proton irradiation has shown particular promise in generating defects that enhance flux pinning capabilities in REBCO films, including YBCO [12,13].

In addition to particle irradiation, two-dimensional patterning of pinning centers has demonstrated significant potential for enhancing superconducting properties, particularly J_c and flux pinning efficiency. Creating a controlled array of defects or pinning centers can significantly improve the material's ability to trap and pin magnetic flux lines. This enhanced flux pinning mitigates vortex motion, thereby maintaining the superconducting state and resulting in improved performance under applied magnetic fields [14–16]. Such patterning effectively increases J_c, making these materials more suitable for demanding applications, including fusion energy systems.

Ion irradiation offers a unique means of combining the benefits of APCs with two-dimensional patterning. Various methods have been developed to achieve these patterns, such as the use of slightly defocused beams in helium-ion microscopes, which create structured arrays of defects [17]. Another effective method involves the use of masks with defined micro-scale patterns during irradiation. By confining ion exposure to specific regions of the YBCO film, this approach creates defect arrays that are highly concentrated in the exposed areas, enabling a straightforward and efficient way to enhance flux pinning [13,18,19]. The use of such techniques represents a significant step forward in optimizing the performance of YBCO films for applications in high-field environments.

Building on this foundation, the present study investigates the effects of 60 keV proton irradiation on the superconducting properties of YBCO films, with a particular focus on the potential benefits of employing micro-scale masks to create controlled defect patterns. These masks, featuring micro-sized holes, are hypothesized to enhance the in-field performance of YBCO films by introducing artificial pinning centers in a controlled and organized manner. By examining the interactions between proton irradiation defect engineering, this study aims to provide critical insights into optimizing YBCO films for fusion energy applications, where the demand for high current densities and strong magnetic fields presents a formidable challenge.

2. Methodology

2.1. Sample Preparation and Irradiation

The YBCO film samples, doped with $BaTiO_3$ nanodots to create vortex pinning landscape, were prepared for the study. BTO-doped YBCO films were prepared via low-fluorine TFA-MOD. A precursor solution (Y:Ba:Cu:Ti = 1:2:3:0.06) was spin-coated on $LaAlO_3$ (001) and heat-treated. Pyrolysis occurred at 650 °C in humid O_2, crystallization at 820 °C in humid Ar with 200 ppm O_2, and annealing at 500 °C in dry O_2. The films had a thickness of approximately 1500 nm on $LaMnO_3$ with silver protective layer of 0.1–0.2 μm. Proton irradiation was carried out at room temperature using a 1.7 MV tandem accelerator equipped with a Negative Ions by Cesium Sputtering (SNICS) source, manufactured by National Electrostatics Corp (NEC) and located at the University of Houston. The irradiation was performed with 60 keV protons at a fluence of 1×10^{16} ions/cm^2 along the c-axis of the films. The proton beam had a diameter of approximately 3 mm, covering all three samples by scanning them.

To ensure targeted and selective irradiation, the samples were masked with transmission electron microscopy (TEM) grids prior to irradiation. Two different types of TEM grids were employed to investigate the effects of micron-scale patterning on the irradiation process. Mask 1 featured hexagon holes with a diameter of 11.5 μm, while Mask 2 contained square holes with dimensions of 90 μm per side. These masks allowed for selective proton exposure of the YBCO films, creating distinct irradiated and non-irradiated regions.

The proton stopping range in YBCO was calculated using Stopping and Range of Ions in Matter (SRIM) software 2013 and determined to be approximately 350 nm from the film surface. This ensured that the irradiation primarily affected the upper region of the films, which was critical for assessing the structural and superconducting changes induced by proton irradiation. Both masked and unmasked regions were subjected to identical irradiation conditions, ensuring consistency across all experimental variables.

Figure 1 illustrates the experimental setup and sample preparation process. Figure 1a shows the NEC tandem accelerator and its SNICS source used for proton irradiation. Figure 1b,c depict the YBCO samples being masked with TEM grids made from copper: Mask 1 with hexagon holes (hole width 11.5 μm, bar width 5 μm) and Mask 2 with hexagonal holes (hole width 90 μm, bar width 20 μm). Figure 1d highlights the post-irradiation samples, clearly showing regions exposed to irradiation and those shielded by the masks. These masked and unmasked regions were critical for comparative analysis of structural and superconducting properties. For the masked YBCO samples, the uncovered regions were carefully cut off prior to magnetic properties measurements to eliminate any potential interference. This step ensured that the analysis focused on the regions of the samples that were either irradiated through the mask or shielded by it, allowing for a precise comparison of the superconducting properties.

2.2. Magnetic Property Measurements

To assess the effects of irradiation on the superconducting properties of the YBCO films, magnetization measurements were conducted using a Quantum Design Magnetic Property Measurement System (MPMS). The measurements included both temperature-dependent magnetic moment (M-T) and magnetization hysteresis loops (M-H) at fixed temperatures.

- M-T Measurements: The temperature-dependent magnetization was measured under an applied magnetic field of μOe, aligned parallel to the c-axis of the samples. This field was chosen to probe the T_c and evaluate the impact of irradiation on the superconducting transition behavior of the films.
- M-H Measurements: The magnetization hysteresis loops were recorded under magnetic fields up to 7 T, applied parallel to the c-axis (H//c-axis) of the samples. These

measurements were performed at a series of fixed temperatures: 10 K, 30 K, 50 K, 60 K, 70 K, and 80 K. The M-H data provided insights into the flux pinning properties and the overall superconducting performance of the films.

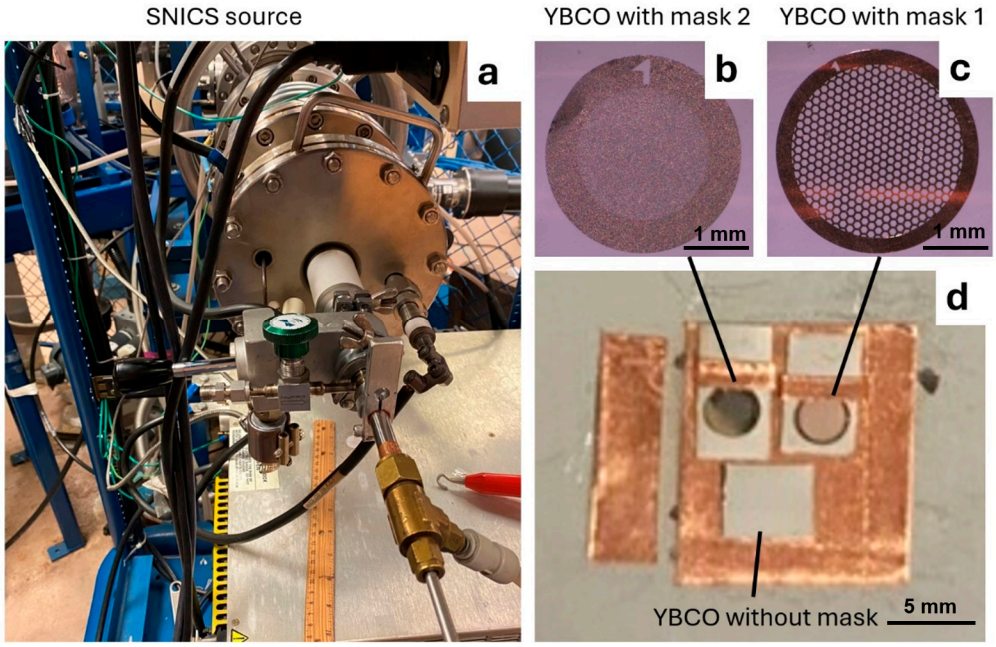


Figure 1. (**a**) The NEC tandem accelerator SNICS source at University of Houston used for the proton irradiation. (**b**,**c**) YBCO samples masked with two types of TEM grids: Mask 1 with 11.5 μm holes and Mask 2 with 90 μm square holes. (**d**) YBCO samples, with and without masking on stage.

The combination of M-T and M-H measurements allowed for a comprehensive evaluation of the superconducting properties of the YBCO films, with specific focus on the differences between the masked and unmasked irradiation conditions. The experimental data provided a deeper understanding of how micron-scale patterning during irradiation influenced the structural and magnetic behavior of the films.

3. Results

Figure 2 illustrates a precise method for selectively irradiating a YBCO film using proton radiation and a micron-scale TEM grid mask. This approach enables controlled defect engineering in superconducting materials, critical for exploring their microstructural and functional properties.

The left panel depicts the experimental setup. A high-energy proton beam, represented by blue spheres, is directed moving toward a YBCO film deposited on a stable substrate. The substrate provides mechanical stability, ensuring the film remains intact during irradiation. Above the film, a yellow TEM grid with rectangular holes is positioned as a mask. This grid allows the proton beam to selectively pass through its openings, targeting specific regions of the film while shielding the rest from radiation. The right panel demonstrates the results of the irradiation. The exposed regions, aligned with the TEM grid's openings, develop localized radiation damage zones, represented by red cubes. These zones arise from the interaction of protons with the YBCO crystal lattice, introducing controlled defects. This process creates a patterned grid-like structure with alternating irradiated and pristine areas, reflecting the geometry of the TEM grid.

This method provides a unique opportunity to investigate the impact of defects on key superconducting properties, such as the J_c and vortex pinning. By systematically varying

the defect distribution, we can study the YBCO film's magnetic behavior under specific conditions, such as high magnetic fields or operational stresses.

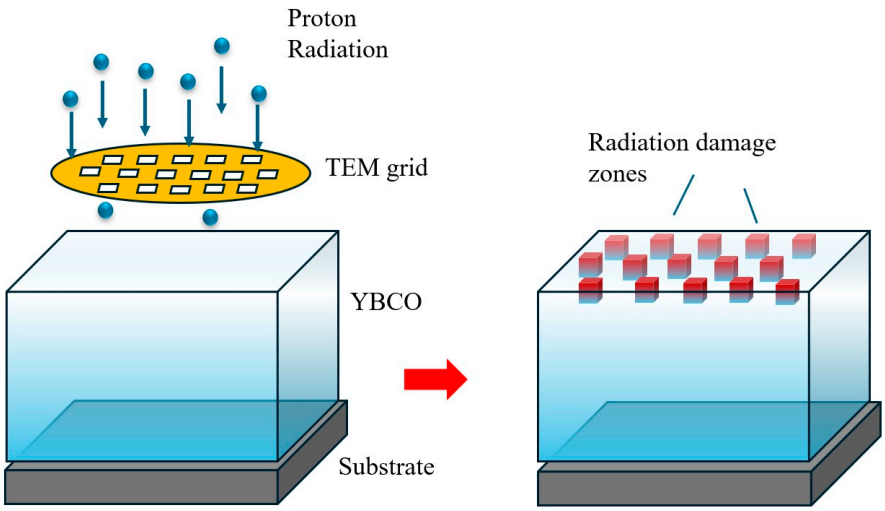


Figure 2. Schematic of proton radiation on YBCO Film using a micron-scale TEM grid mask.

Figure 3 illustrates the temperature dependence of zero-field-cooled (ZFC) magnetization (in units of 10^{-2} emu cm^{-2}) for pure YBCO, unmasked proton-irradiated YBCO, and YBCO irradiated with two different micron-scale masks (Mask 1 and Mask 2) under a low magnetic field of 20 Oe. Across all samples, the superconducting T_c remains consistent at 88.7 K, confirming that the irradiation process, whether masked or unmasked, does not alter the onset of superconductivity.

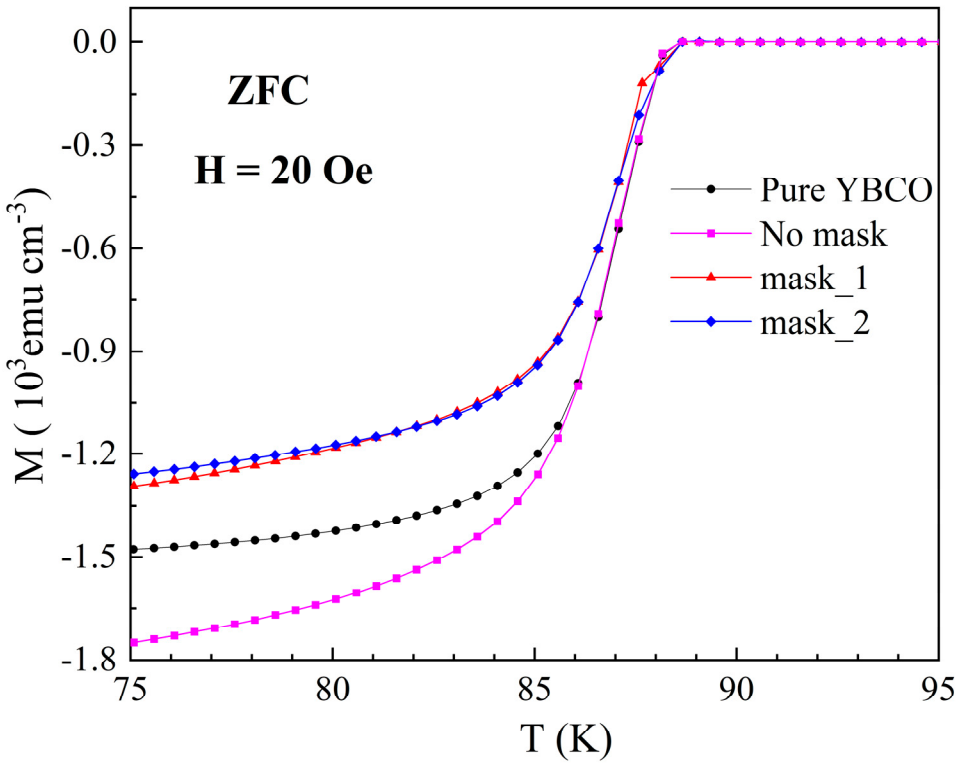


Figure 3. Impact of proton radiation on magnetization and superconducting transition temperature T_c in YBCO films with and without micron-scale masks. M as a function of T under a 20 Oe magnetic field for pure YBCO, unmasked irradiated YBCO, and irradiated YBCO with two different masks (Mask 1 and Mask 2).

The unmasked irradiated YBCO sample exhibits the highest ZFC magnetization in the superconducting state. The masked irradiated samples show the largest reduction in ZFC magnetization, with values nearly identical, highlighting the adverse effects of irradiation-induced defects. The pure YBCO samples demonstrate intermediate behavior, serving as a benchmark for performance. This suggests that the reduction in magnetization is not influenced by the choice of mask design.

This comparative analysis underscores the complex influence of proton irradiation on the superconducting properties of YBCO films. While irradiation introduces beneficial pinning centers to improve J_c, it simultaneously reduces ZFC magnetization. The use of masks during irradiation is expected to help control defect distribution; however, it negatively impact on superconducting performance, as it cannot fully preserve the pristine properties of YBCO. These observations are essential for optimizing irradiation techniques to enhance the applicability of YBCO films for fusion energy applications.

Figure 4a displays the magnetic hysteresis loops at 70 K for pure YBCO, unmasked irradiated YBCO, and YBCO films irradiated with Mask 1 and Mask 2, with the magnetic field aligned parallel to the c-axis (B // c-axis). Among these, the unmasked irradiated YBCO sample exhibits the largest hysteresis loop, indicating stronger flux pinning compared to other samples. Conversely, the samples irradiated with Mask 1 and Mask 2 exhibit significantly smaller hysteresis loops, implying a reduced flux pinning capability introduced by the micron-scale masks.

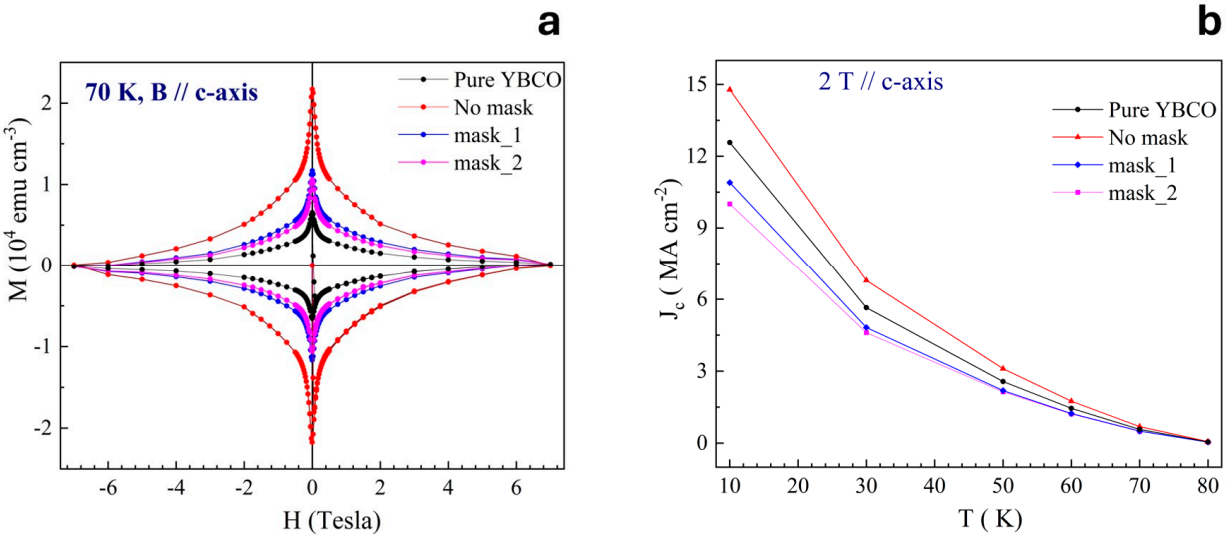


Figure 4. (**a**) Magnetization and (**b**) critical current density of different YBCO films: pure YBCO, unmasked irradiated YBCO, and irradiated YBCO with two different masks (Mask 1 and Mask 2).

Figure 4b presents the temperature dependence of the J_c measured at 2 Tesla, with the magnetic field aligned parallel to the c-axis (B // c-axis). The J_c was calculated using the extended Bean model, based on the formula $J_c = \frac{20\Delta M}{a\left(1-\frac{a}{3b}\right)}$ where 'a' and 'b' (in cm) represent the width and length of the samples, respectively, with the condition that b $\geq$ a. ΔM (in emu/cm^3) denotes the vertical width of the magnetization hysteresis loops.

The temperature-dependent J_c curves demonstrate a consistent decrease in J_c with increasing temperature for all samples. The unmasked irradiated YBCO sample shows an enhancement in J_c compared to pure YBCO, consistent with many reported studies on proton irradiation for flux pinning, while the samples irradiated using micron-scale masks (Mask 1 and Mask 2) show a reduction in J_c compared to unmasked irradiated YBCO. This

reduction suggests that the introduction of defects using masks did not achieve the desired enhancement in J_c but rather compromised the superconducting performance.

These results underscore the complexities in optimizing proton irradiation for YBCO films. While unmasked irradiation can improve flux pinning and enhance J_c, the application of micron-scale masks appears to introduce challenges, reducing the efficacy of defect-mediated flux pinning under high magnetic fields. This highlights the ongoing challenge of maintaining high J_c in YBCO films subjected to defect-engineering techniques.

4. Discussion

4.1. Defect Characteristics and Distribution

The reduction in J_c, rather than its enhancement, following proton irradiation arises largely from the characteristics and distribution of the induced defects. The size, density, and spatial arrangement of these defects are critical factors in determining their efficacy as flux pinning centers. Effective pinning occurs when defects are appropriately matched to the vortex lattice, providing anchor points to immobilize magnetic vortices and enhance J_c. However, the defects introduced by proton irradiation often display significant variability in these parameters, limiting their utility as pinning centers.

Research highlights the importance of precisely introducing defect structures to optimize flux pinning. For instance, B. Maiorov et al. [20] demonstrated that random barium zirconate (BZO) particles, combined with splayed columnar defects, can synergistically enhance in-field J_c when their structure is carefully engineered. Similarly, M. Muralidhar et al. introduced that the improved flux pinning at high fields (14 T) is due to formation of a nanosale lamellar [21]. In contrast, Jooss et al. [22] demonstrated that while well-aligned and sharply defined antiphase boundaries (APBs) can serve as effective correlated pinning centers, extended or poorly structured defects with increased disorder lead to a significant reduction in J_c. Their magneto-optical and microstructural analysis revealed that improper defect morphology and lack of uniformity weaken vortex pinning efficiency. These findings underscore the necessity for precise control of defect structure and alignment to optimize flux pinning performance in YBCO films.

Additional complications arise from secondary effects of irradiation, such as poetical heating caused by mask is in close contact with YBCO or residual stress within the superconducting matrix. These changes can alter the microstructure, potentially degrading superconducting properties further. For example, stress-induced distortions or matrix modifications may interfere with the coherence of the superconducting phase, exacerbating the impact of mismatched defects and contributing to the overall reduction in J_c.

These observations underscore the complexity of the relationship between defect structures and superconducting properties. While proton irradiation holds potential for defect engineering, the current limitations in controlling defect size, density, and distribution present substantial challenges. Innovations such as self-assembled defect networks, nanoparticle doping, or pre-patterned substrates may offer new strategies to address these issues. Such approaches could create more uniform and aligned defect structures, improving flux pinning efficiency while minimizing adverse effects on the superconducting matrix.

Further research is required to better understand the mechanisms underlying these effects and to refine irradiation processes for enhanced defect control. This work is essential for advancing high-performance superconductors, particularly for demanding applications such as fusion energy, where maintaining high J_c under intense magnetic fields is critical. Bridging these gaps will enable the practical application of proton irradiation as a tool for optimizing superconducting materials.

4.2. Impact of Masking Techniques

The use of masks during irradiation aims to control the spatial distribution of defects, theoretically enabling more precise engineering of the flux pinning landscape [17]. This approach seeks to enhance the J_c by creating defects that can immobilize magnetic vortices effectively while maintaining the integrity of the superconducting matrix. However, the experimental results indicate that this method may fall short of producing the optimal conditions necessary for J_c enhancement.

One limitation lies in the inability of the masks to create the uniformity or appropriately scaled defect structures needed to maximize pinning effectiveness. In ion–solid interaction physics, the phenomenon of lateral straggling of low-energy ions poses a fundamental challenge, as it inherently limits the precision of defect placement. As highlighted by earlier studies [23], lateral straggling introduces variability in the spatial distribution of defects, undermining the accuracy of devices fabricated through ion implantation with lithographic masks. This effect is particularly problematic in superconductors, where the critical balance between defect size and density T_c governs flux pinning behavior. Additionally, the complexity of defect morphologies arising from ion–solid interactions, such as cascade overlaps or interstitial clusters, may further reduce their efficacy as pinning centers.

The micron-scale masks used in this study—Mask 1 with 11.5 μm holes and Mask 2 with 90 μm holes, as depicted in Figure 1—were intended to spatially organize the defect landscape. While the masks provided some spatial control, they may not have generated the defect density or pinning strength required to significantly enhance J_c. Instead, they might have introduced defects that were too sparse or lacked the necessary coherence with the vortex lattice. Certain defect configurations may have disrupted the superconducting pathways, contributing to a net reduction in performance rather than the desired enhancement.

Furthermore, the scale and geometry of the mask apertures likely influenced the distribution and morphology of the resulting defects. Mask 1, with smaller apertures, might have produced more localized defects, while Mask 2, with larger openings, could have led to a broader but less dense defect distribution. Neither configuration appears to have achieved the delicate balance between defect density, size, and distribution needed to optimize flux pinning. Instead, both masks may have inadvertently contributed to the formation of defects that were too weak to pin vortices effectively or overly disruptive to the superconducting pathways, ultimately limiting their utility in improving J_c. To overcome these limitations, alternative masking materials or hybrid techniques involving nanopatterning and ion implantation could be explored to refine defect landscapes further.

These observations underscore the complexities of employing micron-scale masks in proton irradiation for defect engineering. While the approach offers promise, further refinement is necessary to address the limitations posed by ion straggling, mask geometry, and defect coherence. Future research should focus on improving the precision of defect formation, perhaps through advanced mask designs or alternative ion irradiation techniques, to fully realize the potential of this method in enhancing superconducting performance.

4.3. Consistent T_c and Magnetization Behavior

A key observation in this study is that the superconducting T_c of pure YBCO, as well as unmasked and masked samples, remains consistent at 88.7 K. This stability indicates that proton irradiation primarily affects M and J_c without altering the fundamental superconducting transition. The preservation of T_c confirms that the intrinsic superconducting properties of YBCO remain intact, even with the introduction of defects intended to interact with magnetic vortices.

This result aligns with previous studies on proton irradiation effects in YBCO films, which consistently demonstrate that T_c remains unaffected by low-energy proton irra-

diation. For instance, research shows that while proton irradiation alters the magnetic properties of YBCO, particularly at low temperatures, it does not degrade T_c [24]. In contrast, heavy ion irradiation or high-energy (MeV-level) proton irradiation often causes significant lattice damage [25] or strain in the film [26], leading to a reduction in T_c. These observations emphasize the advantage of low-energy proton irradiation in defect engineering, preserving the superconducting transition while enabling controlled modifications to other material properties.

From a practical perspective, maintaining a consistent T_c while enhancing in-field J_c is crucial for the development of high-performance superconducting materials, especially for fusion energy applications. The stability of T_c across all samples provides a robust foundation for optimizing other parameters, such as vortex pinning strength. This stability suggests that irradiation techniques could focus on fine-tuning defect characteristics to improve pinning efficiency without compromising the superconducting transition. Integrating computational modeling to predict defect behavior and guide experimental designs could further refine this approach.

However, the similar reductions in J_c observed across samples with varying mask designs highlight the challenges in current defect engineering methods. The inability to achieve significant enhancements in vortex pinning underscores the complexity of designing effective defect structures for operational conditions. Localized heating, stress effects, and the limitations of mask geometries may contribute to these challenges.

These observations reveal the intricate interplay between defect profiles and superconducting performance, emphasizing the need for innovative strategies in irradiation processes. With continued research, proton irradiation could become a powerful tool for tailoring superconducting properties, offering a pathway to develop YBCO materials capable of meeting the demanding requirements of fusion energy and other advanced technological applications.

5. Conclusions

This study investigates the effects of proton irradiation on the superconducting properties of micro-scale masked YBCO tapes, with relevance to fusion energy applications. The results demonstrate that proton irradiation on the micro-scale masked YBCO does not enhance the critical current density (J_c). Instead, irradiation introduces defects that significantly reduce J_c, while the critical temperature (T_c) remains unchanged at 88.7 K across all samples. Variations in mask designs, specifically micron-scale hole sizes, yielded comparable reductions in J_c, indicating that the micro-scale masks did not significantly affect defect characteristics. These observations provide valuable insights into the limitations of current defect-engineering approaches for high-temperature superconductors and underscore the need for improved control of irradiation parameters and defect structures to optimize vortex pinning.

Author Contributions: Conceptualization, D.C. and L.D.; methodology, P.K.S.N. and S.K.; software, P.K.S.N. and S.K.; data curation, P.K.S.N. and S.K.; writing—original draft preparation, D.C.; writing—review and editing, L.D.; supervision, D.C. and L.D.; project administration, C.-W.C.; funding acquisition, C.-W.C. All authors have read and agreed to the published version of the manuscript.

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