Analysing neutron radiation damage in YBa$_2$Cu$_3$O$_{7-x}$ high-temperature superconductor tapes

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Abstract

Superconducting windings will be necessary in future fusion reactors to generate the strong magnetic fields needed to confine the plasma, and these superconducting materials will inevitably be exposed to neutron damage. It is known that this exposure results in the creation of isolated damage cascades, but the presence of these defects alone is not sufficient to explain the degradation of macroscopic superconducting properties and a quantitative method is needed to assess the subtle lattice damage in between the clusters.

We have studied REBCO-coated conductors irradiated with neutrons to a cumulative dose of $3.3 \times 10^{22}$ n/m$^2$ that show a degradation of both $T_c$ and $J_c$ values, and use HRTEM analysis to show that this irradiation introduces $\sim 10$ nm amorphous collision cascades. In addition, we introduce a new method for the analysis of these images to quantify the degree of lattice disorder in the apparently perfect matrix between these cascades. This method utilises Fast Fourier and Discrete Cosine Transformations of a statistically relevant number of HRTEM images of pristine, neutron-irradiated and amorphous samples and extracts the degree of randomness in terms of entropy values. Our results show that these entropy values in both mid-frequency band FFT and DCT domains correlate with the expected level of lattice damage, with the pristine samples having the lowest and the fully amorphous regions the highest entropy values. Our methodology allows us to quantify ‘invisible’ lattice damage to and correlate these values to the degradation of superconducting properties, and also has relevance for a wider range of applications in the field of electron microscopy where small changes in lattice perfection need to be measured.

KEYWORDS

Coated conductor, Damage Cascade, Neutron irradiation, REBCO, Transmission Electron Microscopy (TEM)
1 | INTRODUCTION

In future magnetic confinement fusion reactors, superconducting windings will be vital to generate the strong magnetic fields needed to confine the plasma and enable the deuterium/tritium reaction.\(^1,2\) High-temperature superconductor (HTS) materials, YBCO (Y\(_{1.03}\)Ba\(_{2}\)Cu\(_3\)O\(_7\)) and the more general REBCO (RE = Rare Earth) compounds, are being considered for this application because they can support the high magnetic fields that will be experienced in the windings of a fusion magnet even at temperatures well above 4.2 K.\(^3,4\) However, these superconducting materials will be exposed to a flux of high-energy neutrons in service, and it is important to understand how the interaction of these species with the complex crystal structure of REBCO will affect the superconducting properties. REBCO compounds have an orthorhombic, triple perovskite crystal structure with \(P_{mmm}\) symmetry and highly anisotropic lattice parameters. They consist of a layered structure of CuO\(_2\) (super)conducting planes separated by insulating, charge reservoir layers along the \(c\)-axis, and between the CuO\(_2\) planes one-dimensional (1D) Cu-O chains run along the \(b\) direction.\(^5\) REBCO superconducting tapes with very high critical current densities (\(J_c\)) can be manufactured using coated conductor\(^6,7\) to achieve biaxial crystallographic alignment over long lengths. This is crucial because the small coherence lengths of HTS compounds mean that high-angle grain boundaries act as weak links, severely limiting current transport particularly in magnetic fields.\(^8\)

In operation in the plasma-confining magnets of a fusion reactor, the deuterium/tritium reaction will expose the coated conductors to a flux of neutrons with energies up to 14.1 MeV.\(^9\) This will be especially problematic in small tokamak designs where there is not much room for the addition of shielding or neutron-absorbing layers.\(^2,3,10,11\) Previous research on the effect of radiation damage in REBCO materials shows the same general trend for the evolution of \(J_c\) with radiation dose. Except in very low magnetic fields, an initial increase in \(J_c\) at low dose is followed by rapid decrease after a critical dose which depends on the type of conductor, measurement temperature, and type of projectile. The initial improvement in \(J_c\) has been associated with the introduction of additional non-superconducting flux pinning centres,\(^12,13\) and the subsequent rapid degradation due to both the increased rate of flux creep rate (caused by the reduced separation of flux pinning centres decreasing the activation barrier)\(^14\) and the eventual loss of superconducting cross-sectional area,\(^15\) although no reliable method has been suggested to demonstrate or quantify this second effect. The effect of neutron irradiation on superconducting critical temperature (\(T_c\)) has also been studied and showed a linear reduction in \(T_c\) with a slope of approximately 3% per \(1 \times 10^{22}\) n/m\(^2\) fluence is reported,\(^16,17\) but the width of the superconductor/normal transition temperature is unchanged.\(^18\) In contrast, heavy ion irradiation\(^19\) broadens the transition temperature, and the decrease in \(T_c\) can be larger for the same fluence. Whatever the irradiating species, a systematic decrease in \(T_c\) suggests that damage occurs to the whole volume of the REBCO crystal, presumably with an increase in the concentration of superconducting defects,\(^20\) but relatively little is known about the details of this microstructural evolution.

Frischherz et al.\(^21\) conducted a transmission electron microscopy (TEM) study on YBCO single crystals following neutron-irradiation to a fluence of \(8 \times 10^{21}\) n/m\(^2\). Clear evidence of neutron-induced defects was demonstrated using direct comparison of images from pre and post irradiation samples under two-beam (TB) dark field (DF) contrast to image the local strain fields around defects, giving 3–5 nm black/white contrast dispersed through the matrix. It was inferred from the imaging conditions under which the damage was visible, and comparison with samples damaged by heavy ions, that these isolated features were small cascades that were amorphous or highly distorted in nature. They estimated that only 50% of the incident neutrons created a defect visible in these imaging conditions, and also that samples damaged by proton irradiation contained a majority of smaller features they termed interstitial cluster defects. The possibility of the existence of even smaller point defect clusters not visible in the TEM under any imaging conditions is also discussed, and the authors conclude that the observation that a significant fraction of the measured degradation in superconducting properties in both proton- and neutron-irradiated crystals was recovered following annealing at 300°C implies that these small defects are easily annealed out that but the larger cascades remain (as was confirmed by in situ heating experiments). Zandbergen et al.\(^22\) used high-resolution transmission electron microscopy (HRTEM) on crushed particles of large-grained YBa\(_2\)Cu\(_3\)O\(_7\) to show cascade formation as a result of neutron irradiation up to a fluence of \(2 \times 10^{21}\) n/m\(^2\). No amorphous regions were found, but rather a general lattice disorder manifested as darker contrast and changes in the interplanar spacing along the \(c\)-axis. There is some question whether this disorder arose from a rapid reaction of a thin REBCO sample when exposed to humid air\(^23\) or was generated by the collisions of neutrons with the lattice. These kinds of experiment require access to appropriate neutron irradiation facilities, are usually extremely expensive and result in radioactive samples that can be difficult to study. As a result, considerably more studies have been performed using heavy ions as a proxy for
neutrons, accepting that the damage characteristics are different (especially in dose rate) and attempting to account for these differences by adjusting irradiation parameters. TEM studies on REBCO-coated conductor samples damaged using high-energy (10–1000 MeV) heavy ion irradiation\textsuperscript{25–27} report that columnar amorphous defects are created along the trajectory of these highly energetic ions. Kwon et al.\textsuperscript{25} have also shown the formation of secondary double chain layer (DCL) defects in the matrix surrounding the amorphous tracks formed by 1.4 GeV \textsuperscript{208}Pb irradiation. These rearrangements of the local plane stacking arrangement in the unit cell involve the displacement of such a large number of atoms that numerous extra Cu-O planes are introduced into the perovskite structure over distances of 30 nm around the primary track, and may not be very similar to the damage created by neutrons. Frischer her et al.\textsuperscript{28} chose to use 50 and 85 keV Kr\textsuperscript{+} and Xe\textsuperscript{+} ions, with these much lower energies intended to emulate the damage created by neutrons at the higher end of the 14 MeV energy spectrum. Using TEM TB/DF imaging conditions like those in Ref. (21), they were able to demonstrate similar black/white features a few nm in size to those observed under neutron-irradiation. HRTEM also revealed the amorphous nature of larger cascades with a mean size of 4 nm. Van Tendeloo et al.\textsuperscript{29} performed in situ TEM on \textit{GdBa}_{2}\textit{Cu}_{3}O_{7} single crystals damaged by 300 KeV Ne\textsuperscript{+} ions and 100 KeV Xe\textsuperscript{+} ions, and showed defect clusters appearing as local contrast features under DF conditions. HRTEM investigation of the same samples revealed the amorphous core of spherical cascades between 5 and 15 nm in diameter.

So when REBCO samples are irradiated by species that create amorphous cascades, these defects can be detected both by diffraction contrast and by high-resolution TEM techniques, but there is no agreed methodology to define the more subtle lattice damage caused by smaller defects that must exist between these cascades to explain the systematic degradation in \textit{Tc} values found after irradiation by both neutrons and ions. Chudy et al.\textsuperscript{30} used positron annihilation lifetime spectroscopy (PALS) to quantify the density of small negatively charged defects after doses of $1.2 \times 10^{21}$ and $6 \times 10^{21}$ n/m\textsuperscript{2} in bulk REBCO, and found an increasing concentration with irradiation time of both ‘small defects’ and cascades. However, it is difficult to identify the nature of these defects by this technique.

We have explored whether analysis of HRTEM images from the crystalline but damaged matrix regions between the obvious cascades can offer a technique to quantify the degree of damage. This approach is based on applying image analysis techniques developed for other applications. There are several types of scenes (images) we encounter in day-to-day life, and the most researched type is often called a \textit{natural scene}, namely, an image that was not artificially produced. These natural scene images can contain statistical properties that change in the presence of image distortions, and these properties may be extracted from different domains such as spatial\textsuperscript{31–33} wavelet\textsuperscript{34} and discrete cosine transformations (DCT),\textsuperscript{35} and then used for image quality assessment (IQA) or image compression. However, HRTEM images of crystalline materials are very different from classical natural scenes since they are highly periodic, and this renders most IQA algorithms irrelevant for the analysis of this kind of image.

Shannon Entropy\textsuperscript{36} is a key measure in the field of Information Theory. The amount of ‘information’ represented by a system is equivalent to the amount of uncertainty about its outcome. Shannon called this average uncertainty \textit{entropy}, where the uncertainty in a system is defined by the probability distribution, \textit{p(x)}. This kind of entropy can be used in order to evaluate the amount of information (or equivalently, uncertainty) present in an image. Although the concept of ‘information entropy’ was first coined by Shannon, since then other forms of entropies have been introduced,\textsuperscript{37} all measuring the amount of information contained in a signal.\textsuperscript{38}

Liu et al.\textsuperscript{39} defined the so-called Spatial-Spectral Entropy-based Quality (SSEQ) index, which measures the entropy values of $8 \times 8$ blocks of pixels taken from reference and distorted images in both the spatial and DCT domains, in order to assess image quality compared to a human subjective score. Gu et al.\textsuperscript{40} based their approach on the assumption that a higher contrast is manifested in more valuable information present in an image, which is equivalent to better image quality. For quality evaluation, they applied entropy calculations in the spatial domain after deducting the predictable data from an image using a semi-parametric model. Gabarda and Cristóbal\textsuperscript{41} used a normalised pseudo-Wigner distribution (PWD),\textsuperscript{42} which is an approximate form of the probability distribution function (PDF), but computed in 1 D. This way, the entropy is a directional parameter that can be used as a measure of anisotropy and thus, they suggest, as a proxy for image quality. The majority of Image Quality Assessment (IQA) studies use a database of natural scene reference images and their distorted forms, such as LIVE IQA.\textsuperscript{43} Each of the distorted images is provided with a Difference Mean Opinion Score (DMOS), which is simply a human subjective quality score of an image. This allows the researchers to evaluate their algorithm quality score against a human perspective.

Defining entropy as a measure of complexity in the image, we can now consider how this can be useful in quantifying lattice damage in crystalline materials. To demonstrate the idea of complexity in the Fast Fourier Transform (FFT) domain, consider the case of an image
of a perfect crystalline structure taken in a microscope without aberrations. The FFT domain will consist of frequency components representative of the reciprocals of the lattice spacings, and these components will have intensity distributions that are infinitely narrow Gaussians. As the damage increases two main changes occur: first, the Gaussian intensity distribution of each FFT spot broadens, and second, in the mid-frequency FFT domain, a faint amorphous halo is introduced and the intensity distribution becomes wider. Both effects contribute to the increase in the entropy value extracted from images of damaged samples. A similar explanation can be applied for the DCT domain, although the mathematical representation is different. If local regions of intense lattice damage are created (amorphous collision cascades), they will have entropy values very different to that of the perfect lattice, but more subtle variations in entropy may reveal the averaged influence of lesser degrees of damage to the crystalline matrix, for instance by the creation of DCL defects like those reported by Kwon et al. or even by clusters or arrays of point defects that cannot be detected individually in the images.

This paper reports the results of experiments undertaken to study the microstructural changes in REBCO conductor samples after neutron irradiation, and to show the first attempt to characterise the matrix, as opposed to cascade, damage by entropy-based image quality analysis.

2 MATERIALS AND METHODS

The material used in this study was SuperPower GdBaCO$_2$O$_7$-y coated conductor tape containing BaZrO$_3$ (BZO) artificial pinning centres (APC) and we have compared the as-manufactured and neutron-irradiated states. The irradiation took place in the TRIGA MARK II reactor at the Atominstitut, Vienna, to a cumulative fluence of $3.3 \times 10^{22}$ n/m$^2$. The neutrons have a range of energies from fractions of eV up to 15 MeV where fast neutrons ($E_{\text{kin}} > 0.1$ MeV) comprise 36% of the neutron energy spectrum. After irradiation to this fluence, the $J_c$ value is reduced to about 70% of its initial value measured at 30K in a background field of 15T applied orthogonal to the tape surface ($H \parallel c$), and the critical temperature ($T_c$) has decreased from 90 to 81 K. This decrease in $T_c$ implies that the whole of the sample matrix is damaged, not just isolated volumes (cascades) that do not fully block the cross-sectional transport area of the sample.

TEM cross-sections from both the un-irradiated and irradiated samples were prepared by in-situ lift-out using focused ion beam milling (FIB). Since the superconductor layer is 18 µm below the tape surface, underneath protective coatings of Ag and Cu, the metal layers were first milled away and a layer of Pt was applied directly on the exposed superconducting layer. The preparation of the un-irradiated sample was carried out a ZEISS NVision 40 dual beam FIB using 30 kV Ga$^+$ ions down to a thickness of about 150 nm and final low energy surface cleaning using 2 kV 250 pA Ga$^+$ ions down to about 40 nm with a deliberate wedge shape of the lamella to ensure the ideal thickness for observing microstructural damage somewhere in the sample. For the irradiated samples, the same procedure was carried out in the Materials Research Facility (MRF) at the Culham Centre for Fusion Energy (CCFE) using a FEI Helios FIB dedicated to handling radioactive materials. TEM characterisation was carried out using JEOL JEM-3000F field emission gun operating at 300 kV. Three lamellae from each of the un-irradiated and irradiated samples were investigated using the same sample preparation and imaging techniques to try and rule out artefacts from individual sample preparation processes.

All the 2048 × 2048 pixel HRTEM images (0.013 nm/pixel) were taken at a magnification of 800 K, and filtered using 2D Wiener filtering to reduce noise. We then used the method and MATLAB code described elsewhere to calculate the average spectral entropy values of the four 1024 × 1024 pixel blocks extracted from each image in the DCT domain [for more information on the DCT, the reader is referred to Ref. (50) and the references within]. Further, we expanded the existing code to include entropy calculations from the FFT domain. These two transformations are especially appropriate for the analysis of highly periodic HRTEM images since they represent the spatial frequencies of an image, and thus were chosen for the current analysis. To implement the FFT calculation, we took the absolute values of the complex FFT coefficients after applying a circular binary mask to exclude the low and high-frequency bands (see Figure 1). The mid-frequency band used for the calculation was chosen to encompass the halo present in FFTs from images of the amorphous cores of the collision cascades. The inner and outer radii were selected to be 2.5 and 4.6 [1/nm], respectively. Because we have the amorphous edge region created by the FIB sample preparation process on each of the thin lamellae, we are also able to compare entropy values from images from regions of GdBaCO$_2$O$_7$-y matrix at least partially amorphised by neutrons (cascades), pristine and neutron-irradiated matrix material with values from material from the sample edge that we are confident is fully amorphised by the FIB beam.

A schematic flow diagram of the entropy calculation process is shown in Figure 1.

3 RESULTS

Figure 2A shows a typical HRTEM image taken along the [100]/[010] direction of the irradiated sample. Three different regions can be identified: the crystalline REBCO
FIGURE 1  An illustration of the entropy calculation process. The red circular mask represents the FFT mid-frequency band within which the entropy calculations took place.

FIGURE 2  A cross-sectional HRTEM image of typical amorphous cascades formed in the neutron-irradiated samples and Fast Fourier Transforms (FFT) from selected regions. (A) A HRTEM image taken at [100]/[010] orientation shows three different phases: the GdBCO matrix identified by the periodic layered structure of the ab-planes (1), crystalline BZO rods (2) and amorphous cascades (arrows) identified by the loss of the periodic plane structure. (B)–(D) Fourier Transforms for each phase, (B) for the GdBCO matrix, (C) BZO rod and (D) amorphous cascade, taken from regions 1, 2 and 3, respectively.

matrix (labelled 1), part of a BaZrO$_3$ rod showing characteristic Moiré fringes (2) and approximately circular amorphous regions (3). The two crystalline phases are consistent with the microstructure expected for these SuperPower samples, but the amorphous regions ~10 nm in diameter marked with white arrows look similar to the defects observed by Frischherz et al. after 85 keV Kr$^+$ ion irradiation that were attributed to cascade damage.

FIGURE 3  (A) HRTEM image taken along [100] showing both an amorphous cascade core (5) and a distorted region (6) that can be distinguished from the undamaged crystalline matrix (4). (B)–(D) 7.48 $\times$ 7.48 nm window FFTs taken from regions 4, 5 and 6, respectively. The periodicity of selected regions of the image is demonstrated in the FFTs taken from 9.35x9.35 nm windows, shown in Figure 2B–D, and highlights the crystallinity of the triple perovskite REBCO matrix (Figure 2B) and the cubic BaZrO$_3$ rod (Figure 2C), as well as the lack of order in the amorphous regions in the cascades (Figure 2D). The weak spots in Figure 2D originate from the crystalline matrix at the edge of the amorphous cascade core.

Figure 3 shows another HRTEM image of the irradiated material also taken along the [100]/[010] direction. In addition to the amorphous cascade labelled 5, a distorted region can be seen close to the amorphous core, marked...
6. From the FFT (Figure 3D) and the HRTEM image, this region is more distorted than the undamaged matrix, but not wholly amorphous. This might be the result of a neutron impact at a glancing angle to the direction of observation, or a smaller volume of lattice damage generated by lower PKA that does not extend all the way through the foil thickness. These less severely damaged regions might have been identified as interstitial cluster defects in Ref. (21).

To confirm these amorphous defects are not caused by beam damage in the microscope, sample preparation artefacts or manufacturing defects, we have used the identical characterisation methodology on lamella prepared from un-irradiated samples of the same material. The amorphous regions easily detected in the matrix of the irradiated sample in Figures 2 and 3 could not be found anywhere in three TEM liftouts taken from the un-irradiated reference sample. We can thus assign them with confidence to neutron-induced damage.

Based on these observations, we can estimate the density of large cascades (~10 nm in diameter) in the irradiated material, to be $3.3 \times 10^{19} \pm 1.6 \times 10^{19}$ m$^{-3}$ based on a standard lamella of width of 15 μm and estimated thickness ~40 nm, 1 μm superconducting layer thickness and the observation of 10–30 cascades per lamella. This estimate corresponds to less than 0.01% of the cross section of the superconducting tape being occupied by the presumably non-superconducting, amorphous cascades. Much smaller amorphous regions, and the local disorder in the irradiated samples that is also expected to contribute to the observed degradation in $J_c$ and $T_c$ values, are difficult to accurately quantify using this kind of TEM image as discussed above. The fact that our defect density is much lower than that reported by Frischherz et al. (3.9 $\times$ 10$^{22}$ m$^{-3}$ created by a fluence of $8 \times 10^{21}$ n/m$^2$), stems from the fact that the TB diffraction condition they used is more sensitive to the local changes in diffraction conditions created for instance by smaller interstitial cluster defects. This imaging technique works well for the bulk superconductor crystals free from APCs used by Frischherz et al., but is less suitable for our samples that contain a high density of BaZrO$_3$ (BZO) nanorods that create local distortions due to the lattice parameter mismatch. Conventional lattice images are sensitive to the secondary double chain layer (DCL) defects reported by Kwon et al., and the similar intercalation of CuO$_2$ planes reported Zandbergen et al. in neutron-irradiated powder samples. In Figure 4, we compare typical lattice images of the REBCO matrix in irradiated and un-irradiated samples, but note, as have previous authors, that small defects such as clusters of vacancies or interstitials, and anti-site defects would not be detected.

As mentioned above, formation of a few isolated cascades in a pristine matrix cannot explain the degradation in the superconducting properties of coated conductors at quite low damage levels, so we have explored the use of careful analysis of the HRTEM images to study damage in the matrix even when the triple perovskite lattice is still mostly intact. This damage cannot be quantified directly from the spatial domain, but image transformations such as Fast Fourier Transform (FFT) and Discrete Cosine Transform (DCT) may reveal statistical differences in the frequency domain. Figure 5 shows a scatter plot of the entropy values extracted from regions of more than 500 HRTEM images of pristine non-irradiated REBCO matrix, neutron-irradiated but still crystalline matrix, and amorphous regions. It can be easily seen that the three types of material are very clearly separated according to their entropy values in both the DCT and FFT domains. From the average entropy values from each material, we have calculated the percentage of damage present in the matrix of the neutron-irradiated sample by normalising the average values of the entropies to 0% and 100% in the pristine (assumed to represent the perfect lattice) and amorphous samples (fully damaged), respectively. The average percentage damage values of the neutron-irradiated sample calculated from the DCT and FFT (mid-frequency) domains are 14% and 34%, respectively.

To explore the possibility that entropy values vary according to local conditions of both microscope (such as alignment, drift and aberrations) and sample (such as thickness), we compared the entropy values from the amorphous cluster cores to the fully amorphous region created by FIB damage at the edge of different samples imaged in different microscope sessions. In both cases 512 $\times$ 512 pixels blocks were used instead of 1024 $\times$ 1024 as the amorphous edge regions are rather small, and the data is shown in the inset in Figure 5. We note that, although the numerical entropy values cannot be directly compared with the data taken from larger regions since they scale with number of pixels, it can be clearly seen that the entropy values from amorphous edge and radiation-induced cluster cores are grouped around the same values, giving us confidence in the robustness of the entropy calculations as a tool to assess irradiation damage.

4 | DISCUSSION

Our quantitative image analysis shows that lattice damage leads to more randomness in the image and an increase in entropy values as we progress from the initial
FIGURE 4  HRTEM images taken along [100] and the corresponding FFT images (show in inset) from (A) an as-manufactured and (B) neutron-irradiated sample, showing no obvious intercalation defects in either material.

FIGURE 5  Scatter plot of the entropy values in the DCT and FFT (mid-frequency) domains taken from regions of HRTEM images from pristine, neutron-irradiated matrix and amorphous cascades. Entropy values from amorphous cascades and from the amorphous edge of the TEM lamellae are shown in the inset.

(pristine, but not perfect) REBCO lattice structure to the fully amorphous collision cascades. The source of the difference in the percentage of damage calculated from the DCT and FFT domains may arise from the entropy of the FFT including only the medium frequency coefficients, whereas the entropy of the DCT included all the coefficients. By looking at FFT images of both pristine and amorphous samples, it is clear that the major change occurs around the amorphous halo, that is, more pixels (in percentage terms) undergo change in their intensities, so the entropy was calculated only around the mid-frequency band. Moreover, we used a simple, linear interpolation to calculate the percentage damage in both the FFT and DCT domains, and this may not be appropriate for either or both of these functions. Nevertheless, it can be seen that in both cases, the entropies increase in the irradiated matrix, giving a preliminary quantitative measure of the degree of lattice damage generated by the neutron irradiation in the still highly crystalline matrix.

The effects of neutron-irradiation on the superconducting properties reported by Fischer et al. show a decrease in $J_c$ value at 30 K by about 30% at a neutron fluence of
3.3 × 10^{22} \text{ n/m}^2, after a slight increase at 2.3 × 10^{22} \text{ n/m}^2. The sample we have studied here has been subjected to the highest dose. The critical temperature ($T_c$) decreased linearly with dose from 90 to 81 K and it was speculated that this was a result of the formation of Cu-O di-vacancies along the b-axis.\textsuperscript{30} We have shown here that it is possible to extract from the same samples a measure of damage of a different kind based on quantifying the randomness in HRTEM images, although it does not at this stage give information on the crystallographic nature of this damage. It is interesting to note the similarity of the measured FFT entropy value of 34% (a direct measure of the perfection of the matrix lattice) to the decrease in $J_c$ in this sample. It should be possible to use the same methodology to develop a better understanding of the lattice damage in REBCO by (1) generating a relationship between entropy values and superconducting performance by measuring the entropy as a function of neutron dose, (2) correlating experimental entropy values with those calculated from modelled images containing deliberately added lattice defects of different types and (3) to apply the same analysis to samples damaged by the fast ion irradiation experiments commonly used as a proxy for expensive neutron irradiation campaigns,\textsuperscript{24} and to demonstrate which choice of ion and energy at nominally the same dpa levels gives the most accurate correspondence to the entropy values produced by real neutron damage.

### 5 | CONCLUSIONS

In conclusion, cross-sectional TEM/STEM studies on neutron-irradiated REBCO HTS tapes have shown direct evidence for a high density of amorphous cascades created by neutron irradiation that are not observed in the unirradiated reference samples. We deduce, in line with previous authors, that these rather widely separated cascades alone cannot account for the radiation-induced reduction in $T_c$ because a drop in $T_c$ will only occur when the whole matrix cross section has been damaged so that there is no pristine percolative path through the sample. However, individual lattice defects arising from lower energy PKAs are much harder to detect in TEM images, so we have devised a new statistical approach specifically to quantify how defective the matrix is between the isolated damage cascades. We have shown that neutron irradiation causes an increase in entropy values both in the mid-frequency FFT, and DCT domains as a consequence of local disorder and decreasing lattice periodicity. To the best of our knowledge, this is the first time that this technique has been applied to neutron-damaged REBCO. Future application of this technique to ion-irradiated samples and simulated images with different defect structures may enable the nature of the defects to be elucidated as well as improving our understanding of the best experimental proxies for deducing the rate of neutron damage in a key material for future fusion reactors.

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