**MEASUREMENT OF OXYGEN DISORDER AND
NANO-TWIN MICROSTRUCTURE ASSOCIATED WITH
COLUMNAR DEFECTS IN YBCO***

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MEASUREMENT OF OXYGEN DISORDER AND NANO-TWIN MICROSTRUCTURE ASSOCIATED WITH COLUMNAR DEFECTS IN YBCO

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ABSTRACT

Studies of defects generated by high energy (>1 GeV) heavy ion irradiation in high- T_c superconductors have been performed by transmission electron microscopy (TEM). Our study shows that high dose irradiation leads to the formation of nano-twins, by which the columnar defects are connected. An analysis of the local Fourier components of the image intensity in [001] lattice images indicates that these new "twin" boundaries are much more diffuse than pre-existing twin boundaries in YBCO. The mechanism of the formation of nano-twin boundaries on {110} planes and their possible relation to superconducting properties are discussed.

INTRODUCTION

It is well known that a high critical current density, J_c , can be obtained in high- T_c superconductors only if magnetic flux vortices in the sample are prevented from moving by being pinned on defects. High energy heavy ion irradiation is a well-established method for the controlled introduction of pinning defects in high- T_c superconductors. The ion-induced defects are generally considered to be continuous amorphized columns with cylindrically symmetric strain fields around them. Our previous work showed that the diameters of the tracks actually vary along the c axis depending on the magnitude of the electronic energy loss, and the strain associated with volume expansion can be relaxed by the local oxygen re-ordering (ORO) around the damaged tracks in YBCO [1,2].

Here, we present our recent investigation of the irradiation damage induced by high energy heavy ions by means of transmission electron microscopy. The contrast of the damaged area was studied in both the [001] high resolution image and the zone axis bright field (ZABF)

Fig. 1 Schematic illustration of the high-energy heavy ion irradiation in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$. image, by which a 'nano-twin' network induced by joining oxygen re-ordering regions around the damaged tracks in the orthorhombic YBCO system can be seen. Instead of making a

comparison of full high resolution image series with simulations, we have investigated the application of specific Fourier components sensitive to the structural parameter of interest (in this case the form of the Cu-O chains). The method has been demonstrated to be very effective in the characterization of local changes in oxygen sub-lattice in unirradiated YBCO [3]. Here we examine the relationship between the oxygen content in the c axis projection of a unit cell and the Fourier components in an image associated with the (100) and (010) spacings, and investigate the application of the above approach to the characterization of the form of the O ordering change for the nano-twin boundary in a irradiated high- T_c $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ sample.

EXPERIMENT

Thin ($<20\text{ }\mu\text{m}$) single crystal YBCO samples ($T_c \sim 92\text{K}$) were irradiated with 1.3 GeV U^+ ions at room temperature in the ATLAS heavy ion accelerator at Argonne with the irradiation nearly perpendicular to the c-axis of the specimen. Schematic illustration of the ion irradiation in $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ is shown in Fig. 1. After the irradiation TEM specimens were obtained by ion milling (using $\sim 4\text{ kV}$ argon ion-milling with liquid nitrogen cooling) the irradiated crystal. The depths from top surface of our samples used for TEM examinations here are $\leq 10\text{ }\mu\text{m}$. Calculations using TRIM Monte Carlo energy deposition routine, which show that the mean electronic energy loss of the U^+ ions varies with the depth in YBCO, indicate that the value of the electronic energy losses are approximate constant over the range in depth examined here. The TEM studies were carried out using Philips-CM30 and JEM-4000EXII microscopes operated at 100 and 200 kV.

THE FOURIER COMPONENT APPROACH FOR PRE-EXISTING TWINS IN YBCO

An [001] HREM image of a pre-existing twin boundary, taken near Scherzer focus ($\Delta f = -40 \pm 10\text{ nm}$), is shown in Fig. 2a. The analysis of the oxygen content and ordering near to the twin boundary in Fig. 2a is carried out by obtaining the magnitude of the specific Fourier components of the image intensity of interest for every individual unit cell as described by Walther et al [4]. The intensity distribution in every unit cell in Fig. 2a was thus resampled onto a perfect 16×16 pixels square and Fourier transformed. In the next step the amplitudes $U(\mathbf{g})$ of the Fourier coefficient of the image intensity distribution corresponding to the reciprocal lattice vectors $\mathbf{g}$ were calculated for each of these unit cells.

Figs. 2b-c are grey-level maps showing the Fourier amplitudes $[U(010)-U(100)]/U(000)$ and $[U(010)+U(100)]/U(000)$ respectively for the unit cells from Fig. 2a, together with line scans obtained by vertical projection. The line scan of Fig. 2b shows that the difference between the Fourier amplitudes $U(010)$ and $U(100)$ has nearly the same magnitude for both domains, with the sign being reversed. Thus the changes of the Fourier amplitudes $U(010)$ and $U(100)$ from domain I to domain II as shown in Fig. 2c suggest that we are probing the local changes in the ordering of the oxygen atoms in the Cu-O chains. It is thus significant that the sum of the Fourier amplitudes $U(010)$ and $U(100)$, as shown in Fig. 2c, is drastically decreased locally at the boundary. In fact the drop in Fig. 2c reveals that at a typical pre-existing twin boundary either the average oxygen content is reduced or the oxygen ordering is lost [3]. Furthermore Fig. 2c demonstrates that the width at half maximum of the twin boundary is $1.6 \pm 0.2\text{ nm}$.

Fig. 2 (a) [001] HREM image of a typical twin boundary, (b) $[U(010)-U(100)]/U(000)$ and (c) $[U(010)+U(100)]/U(000)$ two-dimension grey level maps of the Fourier amplitudes from all unit cells in Fig. 2a and their line scans along the twin boundary.

TEM OBSERVATION OF NANO-TWIN STRUCTURE INDUCED BY IRRADIATION

An [001] ZABF image in the samples irradiated with lower fluences of high energy ions is shown in Fig. 3a, in which isolated tracks exhibit lobed "bow-tie" contrast along the *b* axis of the matrix. The contrast behavior is indicative of a transformation of the regions with the re-ordering of the oxygen atoms in the Cu-O chains. The driving force for the local oxygen re-ordering is the strain relaxation. Schematic illustration for the "bow-tie" contrast around the track due to the stress field relaxed by the *a/b* transformation is shown in Fig. 3c.

A remarkable change in the microstructure was observed in the sample irradiated with an ion fluence of $\sim 10^{11}/\text{cm}^2$ (see Fig. 3b). Most of the tracks in Fig. 3b are joined together by the ORO region so that a nano-scale twin network is formed. Schematic diagrams showing how the transformation of approximately parallel sided (110) planar defects might relax the stress between adjacent amorphous columns are shown in Fig. 3d. As indicated in our previous work in low dose irradiated sample [2], around a single track, only the area distributed between the {110} planes (see dashed lines in Fig. 3c) along the *b* axis of the matrix is in favor of the oxygen re-ordering. Thus the regions favoring oxygen re-ordering around the two single tracks can be overlapped in the rectangle shadow region in Fig. 3d. While the ORO region is limited to ~ 10 nm around an isolated track, the maximum ORO distance, L_{max} , between two single tracks can extend up to ~ 50 nm. As the fluence increases, the damaged tracks are closer and the average distance between them becomes small so that a 'nano-twin' network can be formed by the joined ORO areas.

Fig. 3 [001] ZABF images of (a) low dose showing lobed "bow-tie" contrast associated with the isolated columnar defects, and (b) high dose showing a local "nano-twin" network formed by planar defects on $\{110\}$ associated with two or more tracks. Schematic illustrations for (c) the "bow-tie" contrast around an isolated, and (d) how the local rearrangement of the oxygen in the Cu-O chains might relax the stress between adjacent amorphous columns.

ANALYSIS AND DISCUSSION OF THE NANO-TWIN STRUCTURE

An [001] HREM image showing the 'nano-twin' boundaries induced by several damaged tracks close together of the region are shown in the inset of Fig. 4a. The spot splitting of the $\{110\}$ reflections further confirms that the typically diffuse interface observed in Fig. 4a separates regions for which the a and b axes are exchanged and the Cu-O chain directions are re-oriented by 90° . Fig. 4b is grey-level map showing the Fourier amplitudes $[U(010)+U(100)]/U(000)$ respectively for the unit cells in Fig. 4a. As it has been demonstrated above, that no other effects influence the sum of the Fourier amplitudes significantly, the drop in Fig. 4b indicates that at the nano twin boundary the oxygen ordering is lost. Furthermore Fig. 4b demonstrates that the width of the nano twin boundary, over which the oxygen the ordering are at least partially lost, is much wider than the pre-existing thermal twin boundary discussed above. The width of the nano-twins vary from place to place along the boundary, and are approximately are 3~5 nm over all. Since the width of the nano twin, by which the columnar defects are connected in nano scale, is larger than the coherence length in YBCO, one would expect that the nano twin boundaries will affect bulk superconducting properties more significantly than pre-existing thermal twin boundaries..

Fig. 4 (a) An [001] HREM image showing the local 'twin' boundaries induced by damage tracks in the YBCO crystal irradiated with 1.3 GeV U ions. (b) Grey level maps of the Fourier amplitudes $[U(010)+U(100)]/U(000)$ from all unit cell in (a).

It is well established that thermally produced twin boundaries can act as vortex pinning defects, certainly against a Lorentz force in a direction perpendicular to the twin boundary plane. However, twin boundaries can also act as easy channels for vortex motion along the plane of the boundary, both at low applied fields (37-342 G) and low temperatures (20-24 K) as imaged via magneto-optical technique [6]. Interestingly, at low fields and 40 K, the magneto-optical image [6] shows evidence for both the blocking of vortex motion perpendicular to an existing twin boundary, and loss of field from a twin boundary possibly by depinning of vortices. The nano twin boundaries connecting columnar defects, as illustrated in this work, can create an easy channel for vortex motion between columnar defects at ion fluences $\sim 1 \times 10^{11} / \text{cm}^2$. These suggestions apply for low applied fields, while this complex microstructure would be expected to enhance pinning at high applied fields through the action of collective pinning. Our study also suggests that, due to the existence of nano twin network, the twin boundary effect on pinning behavior must be considered in high dose irradiation, even if the single YBCO crystal was detwinned. In addition the nano twin would provide easy "tunneling" of a flux line between two, or several, neighboring columnar pins, as discussed by Nelson and Vinokur [7]. Consequences to a number of published interpretations of such measurements related to pinning and dynamics of magnetic vortices will be discussed elsewhere [8].

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