Correlations Between Critical Current Density, j_c, Critical Temperature, T_c, and Structural Quality of $Y_1B_2Cu_3O_{7-x}$ Thin Superconducting Films.

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ABSTRACT

Correlations between critical current density (j_c) critical temperature (T_c) and the density of edge dislocations and nonuniform strain have been observed in YBCO thin films deposited by pulsed laser ablation on (001) $LaAlO_3$ single crystals. Distinct maxima in j_c as a function of the linewidths of the (00ℓ) Bragg reflections and as a function of the mosaic spread have been found in the epitaxial films. These maxima in j_c indicate that the magnetic flux lines, in films of structural quality approaching that of single crystals, are insufficiently pinned which results in a decreased critical current density. T_c increased monotonically with improving crystalline quality and approached a value characteristic of a pure single crystal. A strong correlation between j_c and the density of edge dislocations N_D was found. At the maximum of the critical current density the density of edge dislocations was estimated to be $N_D \sim 1$-2×10^9 /cm^2.

1. Introduction.

Researchers involved with high-T_c film growth have focussed their efforts on optimizing deposition and annealing parameters to achieve the highest critical current density (j_c) and critical temperature (T_c) [1]. The highest j_c and T_c values have been reported for epitaxially grown films of YBCO, deposited on closely matched lattices such as the (001) faces of $SrTiO_3$ and $LaAlO_3$ single crystals [2]. It is usually assumed that the ultimate values of T_c and j_c in films with a defect-free crystalline structure. However, Siegel et al [3] showed recently that an excellent YBCO film structure may have a relatively low critical current density, close to that of bulk single crystals.

In this paper we present direct evidence that improvement of a film structural quality beyond a specific limit is detrimental for the critical current density in YBCO thin films. Our results indicate that a certain density of structural defects is required for effective pinning of flux lines. Distinct maxima in j_c as a function of $\Delta\omega$ (linewidths of the rocking

curves) and as a function of $\Delta(2\theta)$ (linewidths of the (00ℓ) Bragg reflections) have been found in epitaxial films. The dependence on $\Delta\omega$ implies the importance of edge dislocations in the flux pinning. The dependence of j_c on $\Delta(2\theta)$ indicates that the flux pinning is affected by structural defects responsible for variations in the c-axis length.

2. Film deposition.

The YBCO films investigated here were deposited by pulsed laser deposition from a YBCO target onto the (001) faces of $LaAlO_3$ single crystals (1 x 1 cm^2) using a KrF excimer laser (248 nm). The resulting film thicknesses were about 3000 Å. The details concerning film deposition, annealing and structural characterization are described elsewhere [4]. The films studied exhibited a smooth, lustrous appearance and were free of any visible defects. The critical temperature and the critical current density (at 77 K) were determined by an inductive method [5].

3. Methods of structure characterization.

X-ray structure investigation of the films was carried out on a Siemens D-5000 diffractometer with filtered (Ni) CuK_α radiation. The studies presented in this paper were carried out using θ–2θ X-ray scans and X-ray rocking curves. A brief description of these techniques is presented below.

The Bragg line broadening $\Delta(2\theta)$ is known to arise both from the instrumental resolution as well as from structural properties of the specimen under investigation [6-8]. In our case, the instrumental broadening has been observed to be negligibly small ($\Delta(2\theta)_{instr} = 0.02°$). Over and above this instrumental broadening the line width is usually increased by such factors as small crystallite size and lattice distortions caused by a nonuniform strain in the film due to the presence of edge dislocations, cation substitutions, vacancies, interstitial atoms, etc. [6-8]. In fine-grain polycrystalline samples, the main contribution to $\Delta(2\theta)$ is due to small grain size. In epitaxial films such a contribution is negligible. Thus the major source of the variation in the linewidth $\Delta(2\theta)$ appears to be caused by variations of the c-axis length within the film.

The measured linewidth, $\Delta(2\theta)$, of Bragg reflections exhibiting a Gaussian line-shape can be expressed as [6,7]:

$$[\Delta(2\theta)]^2 = [k \lambda/g \cos\theta]^2 + [2\Delta d/d]^2 \tan^2\theta + [\Delta(2\theta)]^2_{instr}, \qquad (1)$$

where; θ is the Bragg angle of a (00ℓ) reflection, k is a constant (= 0.9), λ is the wavelength of the x-ray radiation (here $Cu_{K\alpha}$), g is the average structural coherence length along the c-axis, d is the lattice spacing in the direction normal to the film surface, and Δd is the variation from the average lattice spacing. $\Delta(2\theta)_{instr}$ is the instrumental broadening. For epitaxial and polycrystalline films consisting of large grains (g $\geq$ 1000 Å) the major contribution to the line broadening comes from the second term [7] and thus the above formula reduces to

$$\Delta(2\theta) = 2 (\Delta d/d) \tan\theta. \qquad (2)$$

For a given Bragg reflection (the (005) reflection was used in this work), $\tan\theta$ = constant, so that $\Delta(2\theta) \sim \Delta d/d = \varepsilon$, where ε is the lattice nonuniform strain [7,8].

It is known that even high quality single crystals consist of sub-micron (~1000 Å) blocks (sub-grains), which scatter x-ray radiation coherently [6-8]. These blocks, forming a *mosaic structure*, are separated by arrays of edge dislocations which are polygonized to form small angle boundaries during the high temperature annealing of the film [9]. Regardless of the film growth mechanism, the merging growth spirals or islands produce planar arrays of edge dislocations which separate mosaic blocks. YBCO films grown by laser ablation usually contain many screw dislocations which were also observed in our films (Fig. 1.).
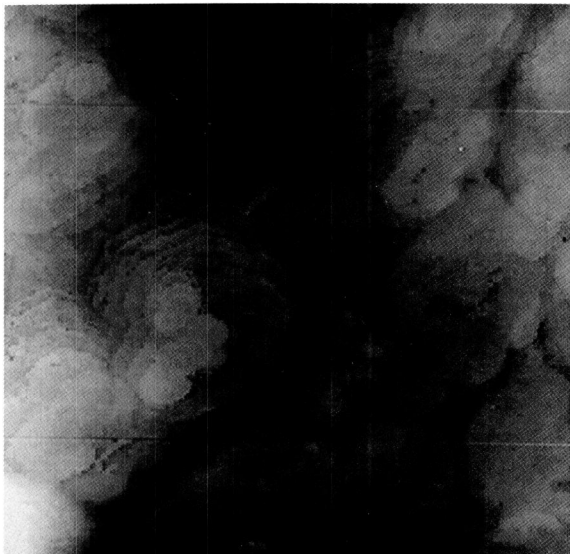

Fig.1. STM image of a laser ablated YBCO film grown on LaAlO3 (100) showing growth spirals. The image covers area of 1.6 x 1.6 μm^2.

HREM studies of YBCO films [10] have shown numerous edge dislocations which occur most likely at the sites where the growth spirals merge, thus creating dislocation walls. These dislocations accommodate the misorientation of the adjacent domains, and are seen as the termination points of the lattice extra half-planes [10].

The density of the edge dislocations can easily be estimated [11]. Assuming that the boundary between two blocks (or islands) consists of a sheet of parallel edge dislocations of the same sign (whose lines are parallel to the a- or b-axis of the film) and if the misorientations of the neighbouring domains exhibit a Gaussian distribution, than the relationship between the measured rocking curve linewidth, $\Delta\omega$, and the density of edge dislocations N_D, is given by [11]:

$$N_D = (\Delta\omega)^2 / 9b^2, \qquad (3)$$

where b is magnitude of the Burgers vector of the edge dislocations. This method is sensitive to dislocations extending at right angles to the crystal surface and compares well with the method relying on etch pit counting [11].

3. Results.

To investigate the influence of structural quality on T_c and j_c, we have studied the mosaic spread in the YBCO films by measuring the widths of the (005) X-ray lines rocking curves. In Fig. 2. the critical current density, j_c, measured in the YBCO films is plotted as a function of $\Delta\omega$, the half-width of the (005) X-ray peak rocking curve. It is evident from Fig. 2, that there is a distinct maximum in j_c vs. $\Delta\omega$ dependence. The maximum is around $\Delta\omega = 0.28^o$.   The density N_D can be estimated here from equation (3) using the Burgers vectors of edge dislocations in YBCO reported by Mannhart et al [12], b[100] and b[010]. For $\Delta\omega = 0.28^o$, the corresponding values are 1×10^9 cm^{-2} and 2×10^9 cm^{-2} respectively. It is worth noting here that these values are very close to the density of screw dislocations reported in Ref. 12. For a small mosaic spread ($< 0.3°$) one edge dislocation is needed to define a single coherent domain, thus the density of edge dislocations should roughly be equal to that of screw dislocations.

For either larger or smaller mosaic spread than $\Delta\omega = 0.28°$, the measured j_c rapidly decreases. While low values of j_c for broad rocking curves, $\Delta\omega > 0.5°$, can easily be accepted, a decrease in j_c for a small mosaic spread is not as obvious. However this result
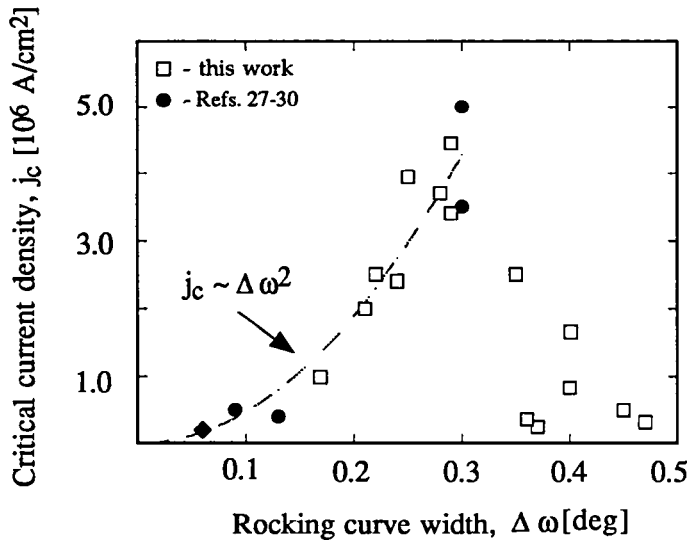
Fig. 2. Critical current density j_c plotted as a function of the half-width of the (005) peak rocking curve, $\Delta\omega$. j_c data for YBCO single crystals (diamond) [18] and a few high quality YBCO films reported in literature [27-30] (full circles) are also shown for comparison.

can be understood if one accepts that an improvement of the film structure, which involves lowering of the density of the edge dislocations, leads simultaneously to substantially weaker pinning of the flux lines. It has been observed [17,18] that YBCO single crystals exhibit j_c values 10-100 times lower than those found in YBCO epitaxial films. As a rule, such single crystals also exhibit very narrow x-ray lines.

All the films investigated exhibited exclusively (00ℓ) reflections of the YBCO compound indicative of a highly c-axis oriented texture. However the full width at half-maximum (FWHM) of the (00ℓ) Bragg reflections varied between films. A similar behaviour in the observed dependence of j_c on $\Delta\omega$ was found in the j_c vs. $\Delta(2\theta)$ diagram, see Fig.3. A plot of j_c vs. $\Delta(2\theta)$, for the (005) peak shows (Fig. 3) a distinct exponential-like decrease in j_c in the range $0.12^o < \Delta(2\theta) < 0.50^o$, with a maximum appearing close to $\Delta(2\theta) = 0.12^o$. The critical current density measured in the films exhibiting narrower Bragg peaks ($\Delta(2\theta) = 0.07$-0.10^o), and thus clearly of a better structural quality, was smaller by a factor of 2-3.

Fig. 3 shows clearly that the lattice strain ε has indeed a substantial effect on the superconductivity in YBCO films, which is in agreement with other observations [13]. Such an effect was also observed for laser ablated and RF sputtered Tl-2212 films as shown in Fig. 3.

4. Interpretation and Conclusions.

Although there is no consensus regarding the role of screw dislocations on flux pinning, a change in the dislocation density leads to substantial changes in jc [12]. We will show here that it is the edge dislocation arrays situated around perimeters of the growth spirals (or islands) which most likely create the centres necessary for the effective pinning of flux lines in YBCO films.
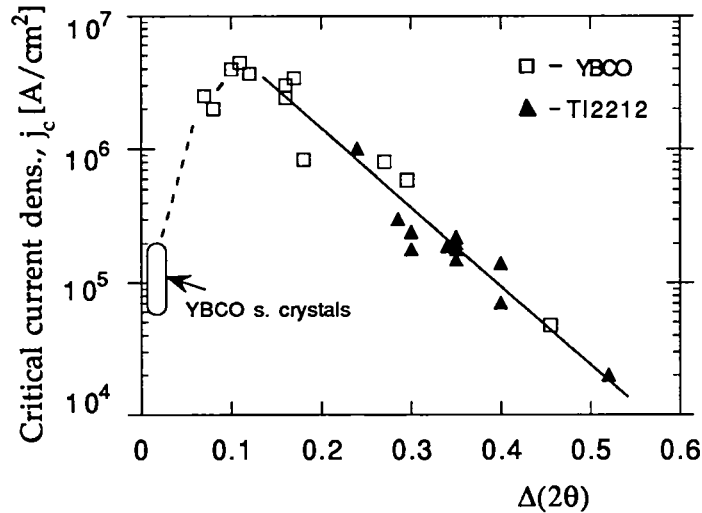
Fig. 3. Critical current density j_c plotted versus half-width ot the (005) Bragg reflection, $\Delta(2\theta)$. For comparison some points obtained for the Tl-2212 films (full triangles) are also presented.

The observed dependence of j_c on $\Delta\omega$ may be explained as follows. Initially, for a large mosaic spread ($\Delta\omega > 0.5^{\circ}$), the dislocation arrays reduce the critical current density relative to the value within the coherent domains by acting either as a partial barrier to the supercurrent flow or as weak flux pinning sites. However, this latter effect has been ruled out by careful analysis of the I-V characteristics of the grain boundaries exposed to microwave radiation [14].

For smaller $\Delta\omega$ ($< 0.5^{\circ}$), the tunneling barrier becomes narrower and a high current density can be transported between the domains. Such an effect should exhibit an exponential-type dependence on barrier thickness since the tunneling current is likely to vary as $j_c \sim \exp(-2a/\xi_b)$, where 2a is an effective width of the barrier between two adjacent blocks, and ξ_b is the coherence length within the barrier [15]. The array of edge dislocations acts as an effective SNS junction. Assuming a direct proportionality between

the pinning potential and the critical current density [16] one can express j_c, as a function of the mosaic spread by

$$j_c \sim U_{eff}[(\Delta\omega)^2/b^2] \cdot exp(-2a/\xi_b)], \qquad (4)$$

where U_{eff} is an effective pinning energy of the edge dislocation. The first term in (4) corresponds to the flux pinning effect due to the interaction between the edge dislocations and the fluxoids, and the second term represents the attenuation of tunneling current as discussed above. For two mosaic blocks of size $L > 1000$ Å and with $\Delta\omega < 0.3°$ the largest width of the tunneling barrier does not exceed $2a = 5$ Å ($2a < \xi_{ab} = 15$ Å), which indicates that a high critical current can flow through the edge dislocation wall almost unperturbed. However for higher dislocation densities, the barrier thickness increases as $2a = \Delta\omega\ L$, and becomes comparable with ξ_{ab}. Hence the tunneling current density falls off rapidly. This behavior is evidenced in Fig. 2 for $\Delta\omega > 0.3°$.
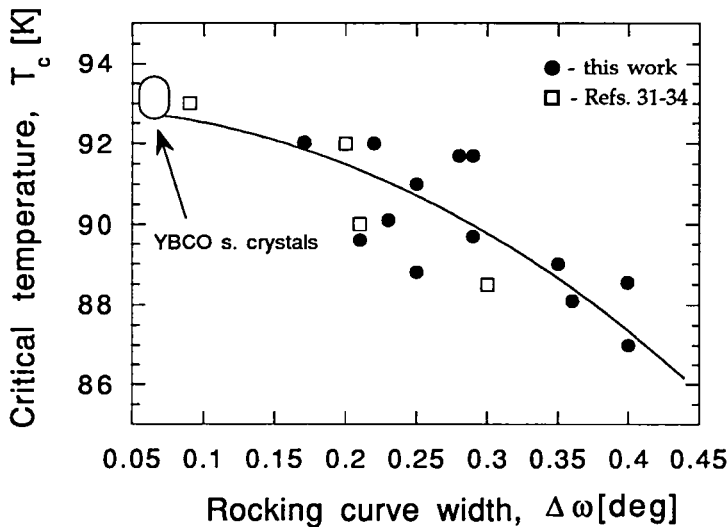

Fig. 4. Ctitical temperature T_c, plotted as a function of the mosaic spread, $\Delta\omega$. As in Fig. 2, the data reported for YBCO single crystals [18] and some of YBCO epitaxial films reported in literature [31-34] are shown (open squares).

The observed increase of j_c in the region of small mosaic spread is proportional to $(\Delta\omega)^2$, see Fig. 2. It is interesting to note that the density of edge dislocations, N_D, is also proportional to $(\Delta\omega)^2$. We suggest that this behaviour is a manifestation of a strong

relationship between the critical current density and density of the edge dislocations in the YBCO thin films. We would like to suggest that the initial increase of the critical current density scales with the density of edge dislocations.

It has already been shown for classical superconductors that the strain field of the edge dislocation is an important factor for the pinning effect [19]. The large local distortion of atomic positions can change electronic properties of the film, creating local inhomogeneities in its superconducting properties. Moreover the dangling bonds of edge dislocations create substantial local electric pertubations in the film, leading to charged grain boundaries. It has been recently postulated that such charged grain boundaries in high-T_c materials should be very effective pinning centres which can attract vortices to the grain boundary more effectively than in classical superconductors [20]. The regions of higher dislocation density act as very attractive sites for flux lines [19, 20].

The presence of dislocation networks (edge and spiral dislocations) creates a nonuniform distribution of the lattice strain which leads to a broadening of $\Delta(2\theta)$ diffraction lines in the films. Therefore it is not surprising that a local maximum in j_c was also found in the dependence of j_c on $\Delta(2\theta)$. A contribution to the scattering/pinning effect from isolated point defects [21] as well as of local aggregations of point defects in the vicinity of dislocation (Cottrell atmospheres) [19], and coherent intergrowths [22], can also be significant.

The role of twin boundaries acting as vortex pinning sites has been recently questioned on the grounds that: 1) the measurements of j_c on the same twinned and detwinned YBCO single crystals showed a negligible role of twin boundaries on flux pinning at low temperatures [23]; 2) in YBCO thin films, observed differences in the twin spacing did not result in changes in the volume pinning force. The microstructure of the twin boundaries did not affect the vortex-pinning behaviour of the film [24]. Moreover, the highest critical currents in YBCO layers were observed in twin-free YBCO/(Nd,Ce)$_2$CuO$_x$ multilayers [25].

The critical current density exhibits a maximum in j_c as a function of the edge dislocation density, while the critical temperature T_c improves constantly with increasing perfection of the film. Fig. 4 shows that $T_c = T_{max}[1 - k(\Delta\omega)^2]$, where k is a constant. One can argue that T_c decreases monotically with an increasing density of edge dislocations, ($N_D \sim (\Delta\omega)^2$). Such a dependence ($T_c = T_c(N_D)$) suggests that edge dislocations are strong scattering centres, whose elimination enhances the critical temperature. This is contrary to the behavior of j_c which requires the presence of lattice defects which act as pinning centres.

This type of behaviour has recently been demonstrated by the introduction of point defects into a high quality superconducting matrix [26]. A few per cent substitution of Pr atoms for Y in YBCO, resulted in a decrease of T_c by about ~2 K, but simultaneously enhanced the critical current density by a factor of 2-3.

Acknowledgements.

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