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On the mechanism of thickness dependence of the critical current density in HTS cuprate epitaxial films

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Abstract. The critical current density, $J_c$, in epitaxial HTS YBCO films strongly depends on the film thickness, degrading from 7-8 MA/cm$^2$ in very thin films (50-70 nm) to less than 1 MA/cm$^2$ in much thicker ones. Electron backscattering diffraction and high-resolution transmission electron microscopy reveal that the nanostructure of pulsed laser deposited epitaxial HTS YBCO films is evolving during growth due to high dislocation mobility. An analysis of existing pinning models leads to a conclusion that any other pinning centres but dislocations can not provide observed $J_c$. Extended linear defects provide a maximum elementary pinning force, if vortices are parallel to dislocation lines, while randomly distributed nanoparticles produce much lower pinning. Thus, the thickness dependence of $J_c$ is determined mainly by the evolution of dislocation structure. Embedding of nanoparticles into HTS cuprates films results in $J_c$ enhancement if extra dislocations are being formed during growth, preserving a high density of dislocations. Nanoparticles reduce the dislocation mobility, fix the dislocation cellular nanostructure and prevent its polygonization.

1. Introduction
Recently high-$T_c$ cuprate conductors (HTS) with high critical current density, $J_c$, for practical applications at 77 K (coated conductors) became a reality. However, a few serious problems still should be solved. The most important ones are (i) to achieve a very high total critical current ($\geq 1000$ A/cm width), very high $J_c$ should be preserved at the coating thickness of at least 2-3 $\mu$m, (ii) to keep high enough $J_c$ $\geq 0.2$ MA/cm$^2$ up to applied magnetic field of at least $\mu_0 H = 5$ T; (iii) to comprehend pinning mechanisms in cuprate films and coatings of any thickness at any applied dc magnetic field and (iv) to find effective methods to enhance the pinning force and the critical current density in HTS cuprate coated conductors. The first problem requires clear understanding what changes occur in the film nanostructure during high-temperature deposition. The dislocation structure is shown to change essentially and rearrangement of dislocations does occur during pulsed laser deposition of thick films at 750-780$^\circ$C [1, 2]. It is shown that a fast $J_c$ decrease with thickness can be suppressed by dividing a thicker film during deposition with very thin barrier layers to get a stack of a few thinner superconducting layers [3]. However, real physical reasons of such an effect still remain unclear. It is shown that a comprehensive relationship between the microstructure evolution and the self-field critical current density decrease cannot be established solely based on degradation of crystallinity [4]. In the present work mechanisms of YBa$_2$Cu$_{3}$O$_{6+\delta}$ (YBCO) film nanostructure evolution with the film growth are studied and analysed on the base of fundamental properties of dislocation arrays in solids. The second problem is very important, since it paves a way to
development of high field superconducting magnets operating at 77 K. It is known that high 
$J_c(77 \text{ K}) \geq 2-3 \text{ MA/cm}^2$ in YBCO films starts to decrease logarithmically at rather low applied fields of 100-200 mT [5]. Embedding of nonsuperconducting dielectric nanoparticles (BaZrO$_3$, Y$_2$O$_3$) appears to be capable of enhancing $J_c$ at higher applied fields [6]. Still unknown mechanisms of vortex pinning reduction can be supposed to exist in HTS cuprate films when they grow up to thick coatings. We are going to clear up a role of dielectric nanoparticles in pinning enhancement as well as to comprehend and to classify possible mechanisms of the effects. Of course, the primary goal to be achieved is finding a new effective way to enhance $J_c$ in thick films and coatings up to 7-8 MA/cm$^2$.

2. The most probable source of $J_c$ degradation in HTS films with the thickness increase

In order to comprehend the real source of $J_c$ degradation in the films and coatings of HTS cuprates with the thickness increase three well known things should be accepted.

(i) High temperature epitaxial growth of a cuprate film on a slightly mismatched (~ 1%) substrate leads to formation of a certain substructure of domains, which are slightly misaligned to each other (< 1º) in $ab$-plane. The domain boundaries form a network of threading edge dislocations $a[001]$, which are parallel to each other and to the $c$-axis of quasi-single-crystal HTS cuprate film.

(ii) Just these linear defects, which total density may exceed $10^{11}$ cm$^{-2}$, provide the strongest correlated pinning of Abrikosov vortices and the highest critical current density in thin cuprate films [5, 7]. This happens due to that the diameter of normal core of such dislocations is about the coherence length in $ab$-plane of HTS cuprate.

(iii) The dislocation substructure develops during the epitaxial growth with formation of low angle boundaries. It appears to be similar to a dislocation cellular substructure described in well known reviews and books (e.g., [8]). Such a structure is subjected to polygonization, which can not be avoided during deposition and, thus, annealing of the film at high temperatures (up to 780-800ºC). The thicker a cuprate film, the longer annealing time.

Polygonization occurs due to not only dislocation slipping but also to their crawling, i.e. thermally activated displacement of atoms at elevated temperatures [8], each dislocation tending to a configuration with the lowest free energy. For example, dislocations of opposite signs can annihilate. Thus, polygonization results in coarsening of dislocation cells. Subboundaries of the cellular structure become more perfect, approaching to an ordinary textured structure with higher and higher angles of boundaries and a low dislocation density. Polygonization obviously results in a decrease of the critical current density in the depositing film, degradation being stronger in layers adjacent to the substrate, since they are longer subjected to a heat. The effect of polygonization is lower on last deposited upper levels of the coating.

The measured thickness dependence $J_c(d)$ for HTS YBCO pulsed laser deposited (PLD) films as well as a few literature points for similar samples on single crystal substrates [9-12] are presented in figure 1. $J_c$ measured in a macroscopic sample first increases with thickness, because very thin films (up to 5-10 nm and sometimes thicker) often are discontinuous. The $J_c(d)$ maximum is observed at $d = 50-70$ nm, when according to our electron backscattering diffraction data the film nanostructure is a so-called “dislocation forest” yet with an average density of about $10^{12}$ cm$^{-2}$ [13]. For thicker films we and other authors found root thickness dependence $J_c(d) \propto d^{-1/2}$ [3, 11, 14].

3. Stabilization of the cellular structure against polygonization by incorporated interfaces

An effective method of dislocation subboundary network stabilization has been found experimentally [3]. It prevents dislocation crawling at high deposition temperatures and thus polygonization of the cellular dislocation substructure and its transformation to an ordinary grained structure due to a long exposure to high temperatures. The method implies introduction of additional artificial oxide interfaces, for example, CeO$_2$.
Figure 1. Summarized thickness dependence of the critical current density. Starlets – our data, squares taken from [9], circles [10], triangles [11], rhombi [12].

However, the nature and the mechanism of this method are not discussed in fact and remain unknown. It should be mentioned that as early as in 1999 we have shown that the critical current density measured by a four-probe technique in 70-80 nm thick PLD YBCO films in a self-field turned out to be 7 MA/cm$^2$ [15]. This seems to be connected with the following: (i) depinning of Abrikosov vortices from dominant pinning centres – linear defects, threading edge dislocations – starts from the surface under the action of Meissner current, when its density reaches the critical value, provided that vortices are perpendicular to the surface, at which the Meissner current flows, (ii) the dislocation structure in thin film ($d \leq \lambda$, $\lambda$ is London penetration depth) is much more rigidly fixed and does not relax during deposition. In order to achieve this in thicker films out-of-plane segments of dislocations should be as short as possible and their ends should be clipped at one or several intermediate interfaces. It is known that the field, when $J_c$ starts to decrease in a parallel field, is [5]

$$H_m^\parallel = 4\pi\lambda J_c(0)/[c \tanh(d/2\lambda)] = 8\pi\lambda^2 J_c(0)/(cd). \quad (1)$$

This means the thicker film, the lower should be magnetic field at the film surface to tear the vortex ends off a dislocation. So, if a thick film would be shared for a few thinner layers then the Abrikosov vortex ends start depinning in the field approximately inversely proportional to the layer thickness. Moreover, the dislocation crawling and, thus, polygonization would be seriously prevented due to clipping of long dislocations on interfaces. Epitaxial interfaces incorporated at the film growth are expected to promote formation of additional $a[001]$ dislocations. Thus, additional interfaces (i) pin a vortex at several intermediate points making it shorter, thus enhancing the field of its ends depinning, (ii) pin dislocations at intermediate points, thus shortening their segments and reducing a probability of their crawling and retarding polygonization, (iii) create a real possibility of generating new out-of-plane $a[001]$ dislocations, which enhance the total pinning force.

A material of the intermediate barrier/buffer layer separating cuprate sublayers should be properly chosen in order (i) to preserve appropriate conditions for the epitaxial growth of the next cuprate coating layer, (ii) to provide required mismatch (0.5-1.0%) between crystal lattices of the HTS film and the intermediate layer and, thus, (iii) to initiate formation of new dislocations and low-angle boundaries by the rotational relaxation mechanism [1, 2].
In this case the density of $a[001]$ dislocations in a consequent cuprate layer would be at least not less than in a preceding one. Therefore, the critical current density would not decease with the coating thickness.

The $J_c$ in cuprate films is shown to be increased also by introducing BaZrO$_3$ (BZO) nanodots. In this study we deposited YBCO films epitaxially using the PLD technique onto various single crystal substrates (LaAlO$_3$, sapphire buffered with CeO$_2$, YSZ). We measured $J_c(H, T, d, \theta)$ and made HREM studies to determine the fine cross-section nanostructure. Actually we detected a certain self-field $J_c$ enhancement with BZO or Y$_2$O$_3$ doping. However, the most exciting and promising phenomenon seems to be self-assembling of BZO nanodots observed recently by several groups [6, 16-18].

4. Self-assembling of BaZrO$_3$ nanodots on $a[001]$ edge dislocations

High-resolution electron microscopic cross-section images of the REBCO films with small additions of BZO are presented in [6, 16-18]. One of the most perfect images made by ORNL group is reproduced from [17] in figure 2.

The observed vertical self-assembling of nanodots is well known especially for semiconductors [19-21]. The theoretical explanation of such a phenomenon was based on preferential nucleation of nanodots in the strain field of buried islands [22].

We suggest the following alternative interpretation of the observed effect. $a[001]$ dislocation cores in HTS cuprates are known [23, 24] to have much more complicated atomic structure than it is usually accepted in the simple continuous model used for description of dislocations in isotropic metal crystals, since cuprates are strongly anisotropic and their structure is layered. Properties of metallic layers in $ab$-plane (CuO$_2$) differ strongly from that of dielectric intermediate layers, such as BaO, as the density of free carriers in the last ones is 3-4 orders lower. In particular, this is the case for the stacking fault energy. The core of $a[001]$ dislocation is extended in $00\bar{1}$ plane and the impurity aggregates at this extended core (the structure of this “nano-island” differs from 123-phase). Taking into account the lattice parameter of optimally doped YBCO cuprate of about 1.17 nm, just the observed spacing should be expected between the nanodots stringed as beads on the cores of dislocations aligned along the $c$-axis and arranged by the rotational relaxation mechanism (figure 2).

5. Stabilization of the cellular nanostructure by embedding of nanoparticles

The self-assembled BZO nanodots formed in YBCO film doped with 1-3% BaZrO$_3$ are disks with diameter of 3-4 nm. Being epitaxially-grown BZO nanodots form colonies like beads. Such a phenomenon was observed in [16-18]. The following questions naturally arise:

Figure 2. Threading $a[001]$ dislocations decorated with BaZrO$_3$ nanodots in HTS YBCO cuprate film deposited using the doped target (courtesy of Dr. Amit Goyal [17]).
(i) How does such an unusual nanostructure of the BZO doped film or HTS cuprate coating appear during deposition?
(ii) Why do the nanodots form colonies stringed on dislocations but not randomly distributed?
(iii) Could this phenomenon promote $J_c$ enhancing in YBCO HTS cuprate in higher applied fields and for thicker coatings?

The first question obviously requires additional studies. However, such an unusual nanostructure can be supposed to be formed due to appearance and growth of $a[001]$ threading dislocations by the rotational relaxation of elastic strains in the growing film [1, 2] as well as due to a high mobility of doping BZO particles. This is the case because of high deposition temperature (750-780ºC). The particles migrating on the growing film surface tend to positions with the highest surface energy, those are exits of dislocation cores to the surface. The core of $a[001]$ edge growth dislocation has strongly distorted quasi-amorphous structure. Accordingly to [23, 24] its diameter in YBCO cuprate film is 3.4-4.0 nm. This seems to be a size of the region with enhanced surface free energy, to which BZO particles approach tending to decrease this energy. The BZO nanodots are seen in the electron microscopy images (figure 2) to have the same order size.

At the further film growth the nanodots (likely disk-shaped) initiate more edge out-of-plane dislocations by the rotational relaxation mechanism. A connected rigid cellular nanostructure of immobile dislocations pinned by nanodots is being formed (figure 3). Such a structure is called a “bamboo structure” [18]. The dislocation nanostructure turns out to be pinned against polygonization and the elementary pinning force increases due to an effective increase of the normal core of decorated dislocations. For relatively large pinning centres the elementary pinning force is known to approach to the limit value $f_{\text{pin}}^\text{max} = \frac{\varepsilon_0}{\xi} \sqrt{2}$ [25]. The dislocation spacing at an optimal doping with nanoparticles can be less than 20 nm. This means that the dislocation density can reach and be stabilized at an extremely high level of more than $10^{12}$ cm$^{-2}$!

![Figure 3](image_url)

**Figure 3.** Stabilization of the dislocation structure and vortex pinning in HTS cuprate coating. (a) Single-layer film with Abrikosov vortices pinned on $a[001]$ edge dislocations. (b) Film with incorporated additional interface. (c) Thick multi-layer HTS YBCO film with rigidly pinned cellular nanostructure. It is stable against dislocation crawling and polygonization due to intermediate interfaces and stringed nanodots.
The works [19-22] concerned rather different 3D cases of self-arrangement with in-plane periodicity specific for the elastic modulus tensor of semiconductors with the cubic symmetry. In the case of YBCO epitaxial films with coherent BZO inclusions the in-plane periodicity is not observed completely, whereas self-arrangement takes place mainly along the c-axis. So, the assumption of elastic energy lowering through alternating growth of BZO islands in the strain field of dislocations and formation of c-oriented edge dislocations in YBCO on coherent interfaces is quite natural for such a system. Nevertheless, a possibility of spatial strain-oscillation effect observed in cubic crystals can not be ruled out in layered materials. It should be noted that non-dislocation self-arrangement mechanism does not contradict to the preferential nucleation of nanodots in strain fields of dislocations, which always exist in epitaxial films. It should be noted also that the dislocation mechanism of nanodots self-arranging concerns mostly laser deposition of HTS films, while film growth during chemical deposition may lead to completely different microstructures and pinning mechanisms [26].

6. Vortex pinning on the dislocation structure stabilized with additional interfaces and nanodots

The primary role of out-of-plane dislocations in the $J_c$ enhancement in thin YBCO films was argued by supporters of strong pinning by 0D defects, such as nanoscale precipitates. The main objection was grounded on the absence of a maximum on the $J_c$ angular dependence at $H \parallel c$ in YBCO films [14]. Such a maximum was expected by the analogy with the case of aligned extended linear and/or planar pins in a bulk superconductor, where it does exist when the direction of applied magnetic field coincides with the direction of correlated defects.

However, it was shown in [5] that the evolution of angle dependences $J_c(\theta)$ with $H$ is absolutely consistent with the model of dominant pinning on edge dislocations ($\theta$ is the angle between the c-axis and applied field). Strongly pinned vortices parallel to the c-axis were shown to exist in tilted low magnetic fields up to a certain characteristic threshold field $H_p$, below which the magnetic induction within the film obeys a simple relation $B = \mu_0 H \cos \theta$. This feature explains the absence of expected maximum at $H \parallel c$ for $J_c(\theta)$ at low enough magnetic fields, whereas the maximum emerges only at $H > H_p$.

On the other hand, advocates of the pinning mechanism by point defects usually appeal to thickness dependences of $J_c$ in YBCO films [14, 27]. Such dependences are essentially different for different film types and YBCO based coated conductors, so do the suggested explanations. Let us consider them briefly. The crossover from $J_c \sim d^{-4}$ dependence to a nearly constant $J_c$ value for the films thicker than 0.6 $\mu$m observed for PLD films was interpreted in the framework of collective pinning model as a crossover between 2D and 3D interaction regimes of the vortex line lattice with a system of weak point defects [27]. Such a theory considers weak random defects (oxygen vacancies) and yields critical current density $J_c = 10^4 J_0$ observed experimentally in bulk single crystal samples ($J_0 = 4 \phi_0 / [3(3\phi_0 \xi)^{1/2}]$ is the depairing critical current density, $\phi_0 = \hbar / (4\pi q_0 \lambda)$ is a typical vortex energy scale, $\xi$ is the coherence length, $\lambda$ is the flux quantum). When strong point pinning centres, such as non-superconducting nanodots with a characteristic radius $r_p$, were considered in [27] in the framework of weak collective pinning theories, the critical thickness of 2D – 3D crossover had been evaluated with more than an order of magnitude discrepancy with the experiment. Moreover, the mean spacing between defects $L$ estimated from the expression $J_c = J_F \xi / (L^2 d^{3/2})$ in supposed 2D regime and from the experimental $J_c$ values gave the absurd result $L = 17$ nm for $r_p = 20$ nm, i.e. the mean spacing between nanodots was found to be less than their mean radius $r_p$. Thus, weak pinning theories are obviously inapplicable for critical current densities in REBCO films.

7. Conclusions

(i) A remarkable evolution of the YBCO cuprate films nanostructure occurs during their epitaxial growth at high deposition temperature due to high dislocation thermally-activated mobility and polygonization process.

(ii) A substantial reduction of the out-of-plane dislocations density and changes of their distribution seem to be a main reason of the $J_c$ dependence on the film thickness.
Effective ways to modify the nanostructure of pulsed laser deposited HTS cuprate films and coatings are described and analysed in order to understand the stabilization of dislocation cellular nanostructure against polygonization and relaxation.

The analysis of pinning mechanisms combined with the nanostructure data leads to a conclusion that incorporated intermediate interfaces and normal oxide nanoparticles may enhance $J_c$ of PLD films due to generation of additional out-of-plane dislocations nucleating on the interfaces by the rotational relaxation mechanism.

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