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Magneto-optical imaging of columnar YBCO films

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Abstract

The columnar growth of superconducting films is a new approach that provides high density of evenly distributed extended defects on the nanometer scale. This growth can be activated by tightly-packed nanoparticles deposited on substrate prior to the pulse deposition of superconducting film.

The magnetization measurements demonstrate that critical current density (J_c) in columnar films is higher than in common epitaxial films. However, in magnetization measurements calculation of J_c is model sensitive. An assumption of ideal critical state frequently proves to be wrong giving incorrect J_c averaged over the entire sample. Missing details of current distribution can be obtained by magneto-optical imaging (MOI) that also allows obtaining local J_c in the samples.

In this paper we report MOI study of previously not imaged relatively thick (about one micrometer) columnar $\text{YBa}_2\text{Cu}_3\text{O}_x$ films in order to clarify if they fit a critical state model and to what extent their properties are modified by the columnar growth. MOI demonstrates a homogeneous penetration of magnetic flux that fits a critical state model. The long-length influence of defects, especially those residing on the edges is less pronounced in columnar than in common epitaxial films. The measurement of the depth of flux front at partial flux penetration allows estimating average low-field J_c , which is in good agreement with extrapolated magnetization measurements. The spatial details of the distribution of magnetic flux provide valuable information for tailoring deposition parameters in order to obtain highest possible value of J_c . MOI could be an ideal tool for measuring J_c when other methods are not available or not appropriate. Using MOI and magnetometry, an important effect of the increase in J_c in aged columnar films has been observed.

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1. Introduction

It is expected that coated high-temperature superconductors (HTS) will find extensive use in cable and magnet applications at liquid nitrogen and liquid hydrogen temperatures. It is attractive to use coated HTS in liquid hydrogen because their critical current density (J_c) there is an order of magnitude higher than in liquid nitrogen. Also, liquid hydrogen is a useful cryogenic liquid that can be utilized as fuel. A synergy of liquid hydrogen and superconductivity would solve many problems of the transition to renewable energy resources [1].

For preparation of coated superconductors, it is important to have a reliable non-destructive method of measuring J_c . A traditional way of doing this is magnetometry [2]. It is however a non-trivial to take into account gradients of magnetic field in the sample. The simple formulae frequently used for calculating J_c work well at high field only. Not knowing detailed information about the distribution of magnetic flux makes calculations of J_c in low magnetic field problematic. A suitable method of measuring J_c in low fields is magneto/optical imaging (MOI) as MOI gives direct information about the distribution of magnetic flux in the sample.

In this work we report MOI of previously not imaged columnar $\text{YBa}_2\text{Cu}_3\text{O}_x$ (YBCO) superconducting films [3,4]. The films were also extensively investigated by magnetometry. An unusual property of significant increase in J_c in aged samples is observed.

2. Experimental

The YBCO films were grown on (100) SrTiO_3 (STO) substrates using an excimer KrF 248 nm laser with pulse duration of 30 ns from an YBCO target. The repetition rate of the laser was 6–10 Hz and the substrate temperature was 780 °C. To achieve columnar growth of YBCO, Au nanodots were deposited in-situ on the substrate by few laser pulses on a gold target [3,4].

The critical current density in magnetic fields along the c axis of YBCO was determined from DC magnetization loops measured on a Quantum Design Magnetic Properties Measurement System (MPMS). For rectangular film of thickness d and planar dimensions a and b ($a < b$), the critical current density J_c was calculated from an averaged magnetic moment (m) using the equation:

$$J_c = \frac{4m}{a^2bd(1 - \frac{a}{3b})} \quad (1)$$

In this equation, it is assumed that the current is constant in any point of the cross sections of the film. The J_c is considered to be field dependent and the gradients of magnetic field are ignored. Due to the gradients, however, this formula is not applicable in low magnetic fields [2]. In low fields, J_c was calculated by an exponential extrapolation from high fields.

Flux penetration into the films was investigated using a MOI method based on the Faraday effect in an in-plane magnetized bismuth-substituted ferrite garnet sensor film. The sample was mounted on the cold finger of a continuous He-flow cryostat. A variable magnetic field was applied perpendicular to the sample using resistive coils. The brightness of MOI image obtained with the crossed polarizers was used to measure local magnitude of perpendicular magnetic flux density. J_c for a square film of the planar size $2a$ was calculated on initial stages of the penetration of magnetic flux using formula [5]:

$$d_p = a/\cosh(\pi H/J_c d), \quad (2)$$

which links J_c , applied field H and the depth of penetration of magnetic flux d_p .

3. Results and discussions

Figure 1 show magneto-optical images of a 0.8 micrometer thick columnar film (a, c) in comparison with control epitaxial film (b, d) at temperature of 70 K and magnetic field of 17 mT (a, b) and 51 mT (c, d). The films were prepared using the same deposition conditions. The low-field images a) and b) show that due to a higher J_c , magnetic field penetrates into columnar film to a smaller depth. The penetration of flux into columnar film is also more homogeneous and less influenced by the defects. The same differences persist in MOI images c) and d) recorded in a higher field. The columnar film also shows a higher roughness of flux front, which is the result of stronger interaction between vortices and defects restricting motion of flux front. Figure 1 emphasizes difficulties in preparation of homogeneous thick YBCO films. The homogeneity, however, is easier to achieve using columnar growth.

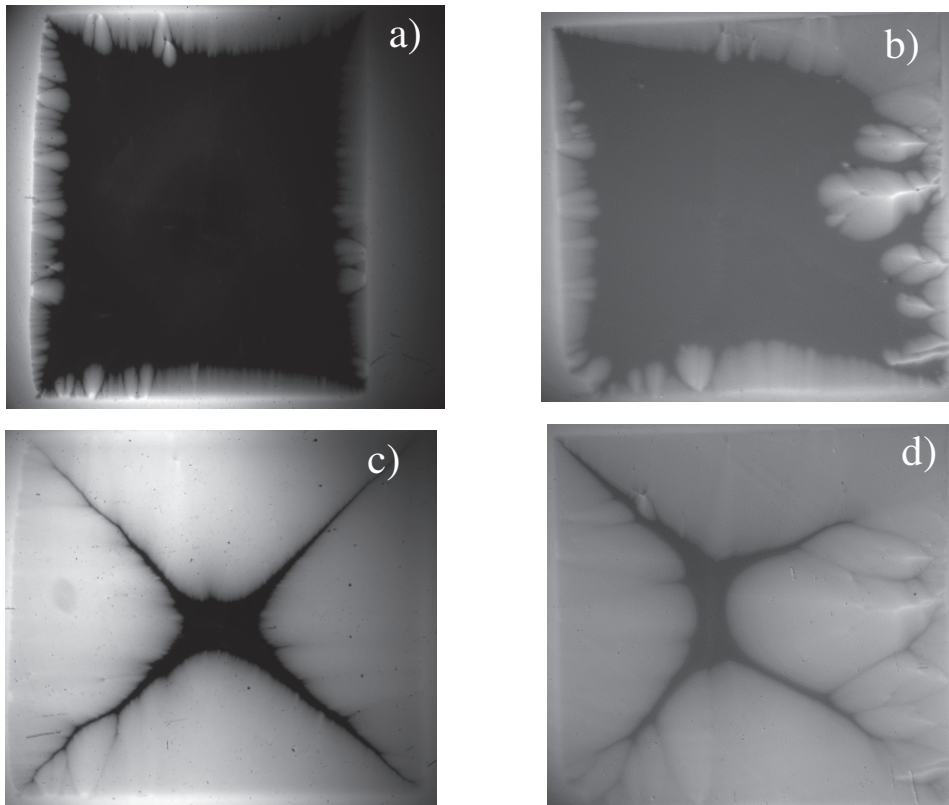


Fig. 1. Magneto-optical images of a 0.8 micrometer thick columnar film (a, c) and control epitaxial film (b, d) at temperature of 70 K and magnetic field of 17 mT (a, b) and 51 mT (c, d). The columnar film shows better homogeneity and higher J_c . It is less influenced by large defects and shows a ragged flux front at high field (c) strongly affected by pinning centers.

The J_c values calculated from magnetization measurements and MOI images using formulae (1) and (2), respectively, are usually in good agreement with each other. It was surprising then to find systematically higher (about 60%) values of J_c in MOI measurements performed on older films stored in desiccators for about three years. After finding this inconsistency, aged films were re-measured in MPMS that allows obtaining higher-field J_c . The results of these measurements are shown in figure 2.

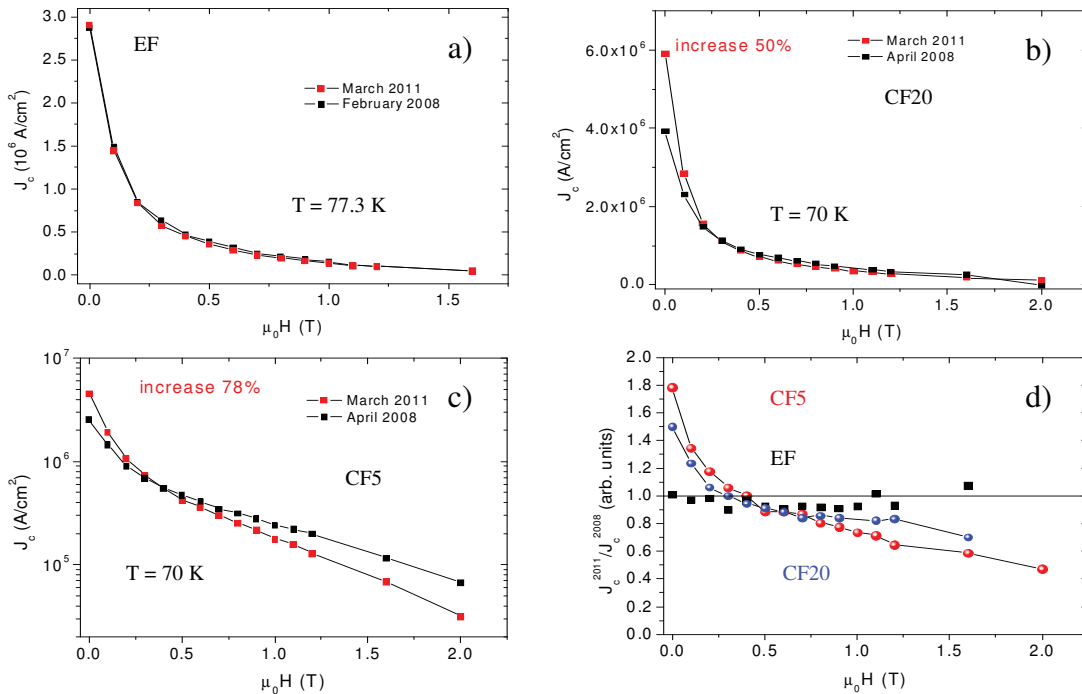


Fig. 2. a) Magnetic field dependence of critical current density for an epitaxial film (EF) from the measurement made in February 2008 (black squares) and in March 2011 (red squares). There is no change in J_c during three years of storage. b) Time evolution of the magnetic field dependence of J_c in a columnar film grown with assistance of deposited by 20 laser pulses Au nanoparticles (CF20). Black squares show measurement is 2008 and red squares in 2011. c) Magnetic field dependence of J_c as measured in 2008 (black symbols) and 2011 (red symbols) for columnar film grown with assistance of Au nanoparticles deposited by 5 laser pulses (CF5). d) Effect of ageing on J_c of the epitaxial film (black) and two columnar films CF20 (blue) and CF5 (red).

Figure 2a is for a common epitaxial film. Black squares show magnetic field dependence of J_c at 77.3 K as measured in February 2008. The red squares are from the measurements in March 2011. There is no change in J_c in this film during almost three years of ageing. Such behaviour is common for properly deposited YBCO films. In contrast, columnar films demonstrate significant increase in J_c at low magnetic fields as shown for the film CF20 in figure 2b, which was grown with the assistance of Au nanoparticles deposited by 20 laser pulses. The increase in J_c in his film is about 50%.

Figure 2c gives another example of $J_c(H)$ for columnar film CF5 grown on Au nanoparticles deposited by 5 laser pulses. This plot is presented in logarithmic scale to clarify changes in J_c in high magnetic field. It shows that increase in J_c in low magnetic fields is accompanied by its decrease in high fields.

Figure 2d summarises all data plotting the ratio of J_c measured in 2008 and 2011. It demonstrates absence of change in J_c in epitaxial film, increase in J_c in columnar films in low magnetic fields and its decrease in high fields.

Such behaviour can be explained by the evolution of defects in columnar films. The columnar films are grown on the substrate densely covered with gold nanoparticles of the diameter of about 20 nm. During the laser pulse these nanoparticles are in the liquid state. They absorb YBCO plume and when start solidifying after the pulse, deposit epitaxially what is absorbed to the substrate below. The repeated pulses add more YBCO layers and push gold nanoparticles up. Below each nanoparticle an YBCO

column grows. The YBCO columns are well connected. In fact the boundaries between the columns are represented by just few dislocations extended from the bottom to the top of the film.

The distance between dislocations is about 20 nm. It is a highly correlated array of extended defects. These defects are important for pinning of magnetic flux. The improvement in pinning was the primary goal of growing columnar films. The enhancement of pinning for magnetic field along the *c*-axis of YBCO, i.e. perpendicular to the film plane or along the extended defects was confirmed by angle-dependent transport measurements [3,4]. Still, these defects form a metastable state that is able to relax. While in common epitaxial films abundance of different varieties of defects [6] and the absence of order prevent their motion, it is likely to happen in columnar films. Our experiment shows that motion of dislocations could take place even at room temperature, although with a very long characteristic time of relaxation of few years.

In the process of the annealing of dislocations, there is a steady decrease in grain boundary misorientations. Since critical current density is very sensitive to the average angle of misorientations, it is not surprising to find an increase in J_c with time. The dislocations, however, are important for pinning of magnetic flux and the decrease in their number negatively affects critical current in high fields as demonstrated in figures 2c and 2d.

The number of dislocations in the sample can be varied by the size and density of gold nanoparticles deposited to the substrate prior to the growth of superconducting film. The smaller number of laser pulses to the gold target (sample CF5) creates more-isolated networks of dislocations, which results in lower J_c , but larger increase of J_c with time as shown in figures 2c and 2d. In contrast, larger number of pulses (sample CF20) leads to a larger J_c but slower increase of J_c with time.

It is worth mentioning that J_c of the films shown in figure 2 is already high enough to be suitable for most applications and is among the best values achieved by other methods. The additional increase in J_c does not require an effort and can be further enhanced by a proper annealing procedure. Having high J_c in zero magnetic fields makes these films especially promising in cable applications. Due to a slow decrease of J_c with magnetic field they are also suitable for magnet applications. However, in this case long term exposition to high temperatures, even room temperature should be avoided.

The MOI images similar to those shown in figure 1 have been recorded in a wide range of temperatures starting with 11 K. The magnetisation measurements have also been performed at different temperatures showing much higher potential of these films for applications in temperatures lower than boiling temperature of liquid nitrogen, particularly in liquid hydrogen.

4. Conclusions

A combination of magneto-optical study and magnetometry allowed to observe an increase in critical current density in aged columnar YBCO films. It takes place due to the annealing of extended defects formed during the deposition of YBCO. The increase in critical current density with time would make columnar films attractive for cable applications and the observed evolution of critical current density in high fields would help to optimize exploitation routines to extend the life time of coated conductors in magnet applications.

Acknowledgements

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