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Features of epitaxial growth of YBCO in the windows of the preliminary topology mask

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This work is devoted to the study of epitaxial YBCO films obtained by laser sputtering during deposition of YBCO into the windows of the special preliminary topology mask. The morphology of the surface of the obtained structures was studied by electron microscopy, the electrical characteristics of superconducting bridges were measured: the critical temperature and the critical current density. The possibility of forming YBCO structures of a given topology with defect-free regions of micron sizes while maintaining high electrophysical parameters of the superconductor is shown. This opens up the possibility of reproducibly forming submicron-scale elements in the created defect-free areas using ion etching or ion implantation.

Keywords: nano- and microstructures, defects, growth in local areas, YBCO.

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1. Introduction

Manufacturing technology of structures based on films of high-temperature superconductor $Y_1Ba_2Cu_3O_{7-d}$ (YBCO) to date reached a high level. However, the problem of obtaining YBCO films or individual elements with a high density of the critical current and simultaneously having a perfect („very smooth") surface is actual. This is due to the fact that YBCO films with better electrophysical characteristics usually contain large structural defects of various types.

The issue of reducing the defectiveness of YBCO films was studied earlier in many papers (see, for example, [1–3]). The defectiveness decreasing and the smoothness increasing of the film surface was achieved by lowering the growth temperature. At the same time, along with the defectiveness decreasing, the electrophysical parameters of the film deteriorated. And here it was necessary to make a certain compromise, namely, to use the films grown at a low temperature with an acceptable morphology, but with reduced electrophysical parameters. At the same time, in such „compromise" films there are still numerous structural defects and precipitates, but with a slightly lower density and smaller sizes. This limits the possibilities of reproducible production of devices with submicron-sized elements that have extremely high characteristics. In the present paper we demonstrate the capabilities of the topology mask (TM) method proposed by us for a comprehensive solution of the problem of creating regions of YBCO film at given substrate locations that have a perfect („very smooth") surface while maintaining high electrophysical parameters of the superconductor. Such predetermined places are extended areas several micrometers wide, which are windows in the topology mask. In such structure, upon YBCO deposition, the gettering properties of TMs appear, leading to the formation along the TM boundary of a region several micrometers wide, which has perfect („very smooth") surface and is completely free from defects observed in a scanning electron microscope. The same perfect region of the YBCO film also appears in the narrow extended windows of the topology mask. In this case, the growth temperature remains high and optimal from the point of view of the electrophysical characteristics of the superconductor film. As a result, a structure is formed in which the YBCO film remains defective over large areas, and in extended TM windows a few micrometers wide the film becomes smooth. Moreover, on large areas and in narrow TM windows, the electrophysical characteristics of the YBCO film are extremely high. This opens up the possibility to reproducibly form the complex circuit topology with submicron elements: first, YBCO film structure is formed with defect-free regions in the required places, and then critical submicron circuit elements are reproducibly created in these places. It is important that the electrophysical characteristics of the critical YBCO elements remain extremely high.

The discussed topology mask method was previously proposed by us for the fabrication of planar structures based on YBCO films. Compared with other methods of forming YBCO-structures, the TM method has a number of advantages, which were demonstrated in the papers of the authors of this article [4–6]. The main feature of the TM method is that the YBCO deposition is carried out at the final stage of the structure topology formation, while a superconducting YBCO film is formed in the TM windows, and separating regions are formed on the TM surface.

In this paper, we present the results of studying the common factors of the defects formation in the film during the YBCO deposition into the TM windows by the laser sputtering method. Particular attention is paid to the formation of film regions that have perfect („very smooth“) surface and are completely free of defects, while maintaining the limit electrophysical characteristics of the film.

2. Technology for YBCO structures formation using the preliminary topology mask method

Single-crystal (r-cut) sapphire substrates 10×10 mm in size were used in the study. Four options of TM material were used:

1. Single-layer TM — „coldCeO_2“ — amorphous cerium oxide $1.5\,\mu$m thick, deposited without the substrate heating. Before formation of this type TM, the epitaxial sublayer of cerium oxide was preliminarily deposited on the entire surface of the substrate.

2. Two-layer TM — „coldCeO_2/hotCeO_2“ — first, a layer of amorphous cerium oxide coldCeO_2 100 nm thick is deposited on the substrate without heating, then — 50 nm hotCeO_2 cerium oxide at the epitaxial growth temperature. When using this type of TM, the epitaxial sublayer of cerium oxide is formed in the windows only.

3. TM „islands“ — in this option, the TM material is the surface of the sapphire substrate, which separates the regions covered with a sublayer of epitaxial cerium oxide. Such „islands“ of epitaxial cerium oxide 50 nm thick on the sapphire substrate were obtained as a result of chemical etching of the structure formed earlier during the YBCO deposition on the substrate with a two-layer TM „coldCeO_2/hotCeO_2“.

4. TM „coldYBCO“ — a layer of amorphous coldYBCO 250 nm thick is deposited on the substrate without heating. Before formation of this type TM, the epitaxial sublayer of cerium oxide was preliminarily deposited on the entire surface of the sapphire substrate

When using all types of TM, the topological pattern was formed by the method of lift-off photolithography carried out on amorphous layers of coldCeO_2 and YBCO deposited without the substrate heating. For photolithography the MJB4 alignment and exposure setup with UV400 optics ($\lambda = 350-400$ nm) was used.

After TM formation, the YBCO layer was deposited on the substrate at the epitaxial growth temperature. As a result, in the mask windows, i.e., on epitaxial CeO_2, the superconducting elements of a given pattern were formed, and between them — isolation regions.

The deposition of the materials used was carried out by laser sputtering. We used the LPX200 excimer laser, KrF mixture, laser radiation wavelength 248 nm, pulse width 27 ns, pulse energy 350 mJ (pulse power 13 MW), repetition frequency 50 Hz. The optical system providing focusing of the laser beam on the target surface consists of a quartz prism and lens with a focal length of 30 cm. The size of the laser beam on the target surface is 1×4 mm. The energy density on the target surface is ~ 10 J/cm. The distance from the target to the substrate is 60 mm. To exclude local overheating of the target and to ensure uniform wear, the rotation and axial movement of the target were used. The target was sputtered in oxygen atmosphere at a pressure of 20 Pa. The growth rate was 0.3 Å per impulse. The substrate was heated by radiation. The heater is a quartz tube with a diameter of 30 mm with a resistive spiral made of heat-resistant stainless steel. The substrate is located inside the heater at a distance of 1 cm from the edge of the heater.

We will call the growth temperature the temperature of the control thermocouple, when its junction, upon preliminary taking the temperature profile in the reactor, is located at the substrate location. Since the heating is produced by radiation, the actual temperature of the substrate differs from the junction temperature of the control thermocouple. Also note that substrates with different types of TM can have different temperatures under formally identical growth conditions.

The epitaxial growth of CeO_2 (001) was carried out at temperature of 950°C; during the deposition of epitaxial YBCO (001) the growth temperature varied from 835° C to 888°C. After deposition process completion, oxygen was admitted into the growth chamber to atmospheric pressure, after which the substrate heater was turned off.

Contacts to the formed structures were made by thermal sputtering. Contact material — silver $100-200$ nm thick.

3. Morphology and electrophysical properties of YBCO structures

The surface morphology of the structures was studied on the CarlZeiss EVO 10 scanning electron microscope (the images were obtained in secondary electrons). The electrical characteristics of the structures were measured by the four-probe method in a Dewar vessel with liquid nitrogen.

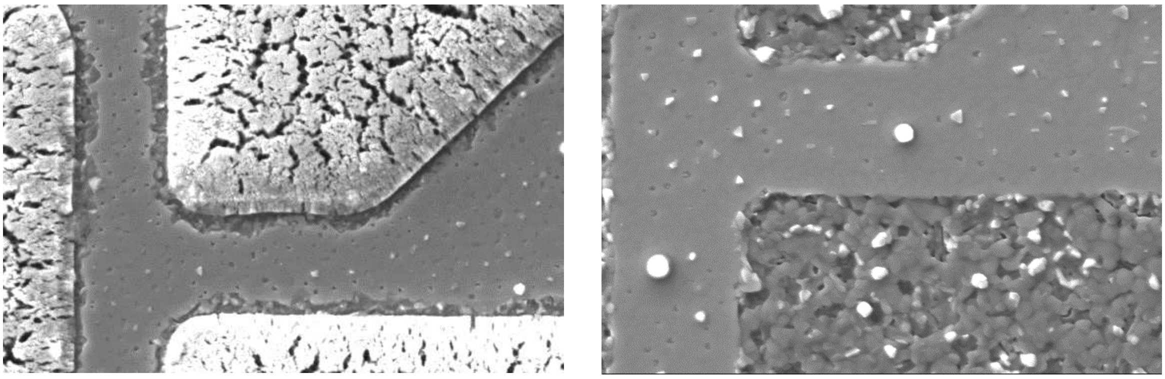
On the array of structures with different options of the TM material considered in this paper, the dynamics of changes in the morphology of the epitaxial superconducting YBCO film with the growth temperature change was traced:

– the presence or absence of various types of defects;

– difference in the density of defects in local regions of micron size and „wide“ regions of the film;

– the influence of boundary TM — YBCO film on the presence or absence of defects of various types near it, i.e., occurrence of the gettering properties of TM, leading to the appearance of a region of a smooth film near such boundary.

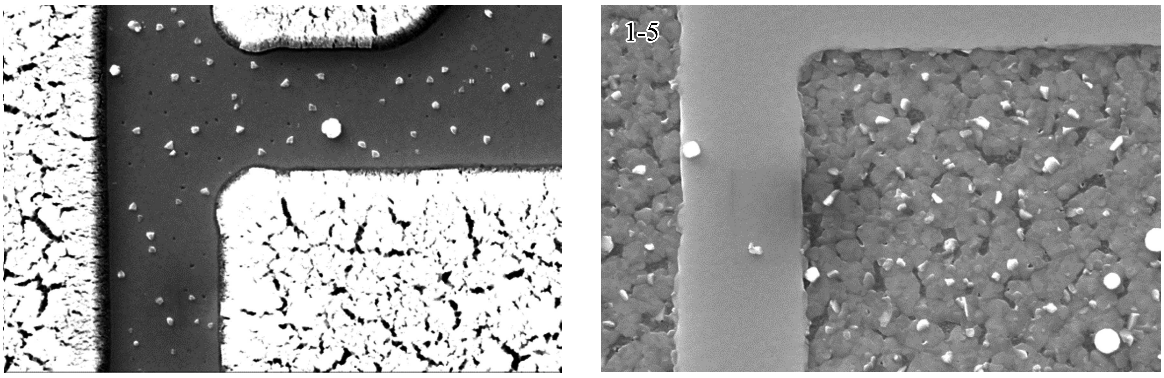
Characteristic changes in the surface morphology of the YBCO film as the growth temperature decreases from 888°C to 835°C are shown in surface photos of



a *b*

— 1 μm L477 (TM "cold CeO_2") — 1 μm L476 (TM "islands")

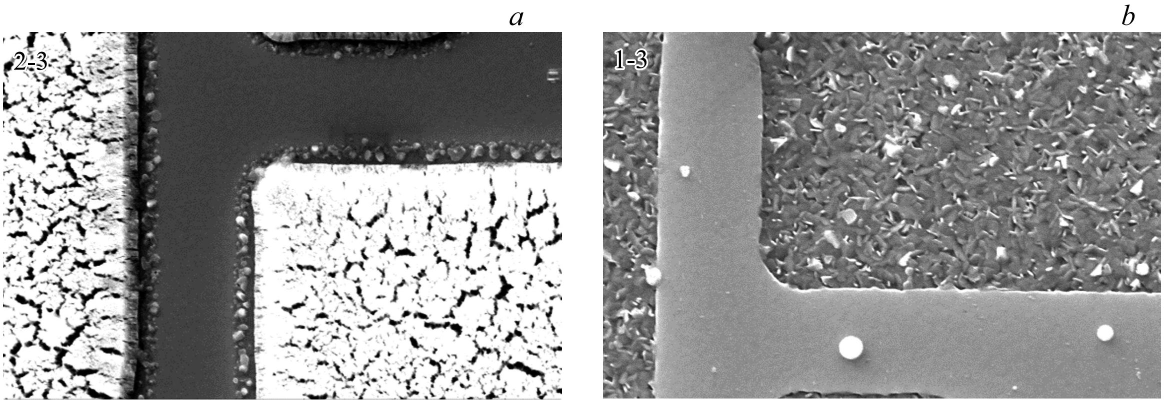
Figure 1. YBCO surface morphology in structures L477 and L476. The structures were obtained at the growth temperature of 888°C. Zoom 50′000.



a *b*

— 1 μm L479 (TM "cold CeO_2") — 1 μm L498 (TM "islands")

Figure 2. YBCO surface morphology in structures L479 and L498. The structures were obtained at the growth temperature of 866°C. Zoom 50′000.



a *b*

— 1 μm L495 (TM "cold CeO_2") — 1 μm L500 (TM "islands")

Figure 3. Surface morphology of YBCO in structures L495 and L500. The structures were obtained at the growth temperature of 851°C. Zoom 50′000.

structures L477, L479, L495, L496, manufactured on the substrate with a single-layer TM coldCeO_2, and structures L476, L498, L500, L501, where the TM was a sapphire surface („island“) (see Fig. 1–4). On structures with other options of TM, the morphology changes approximately in the same way.

Three main types of defects are observed in YBCO films obtained by laser sputtering: CuO precipitates, which

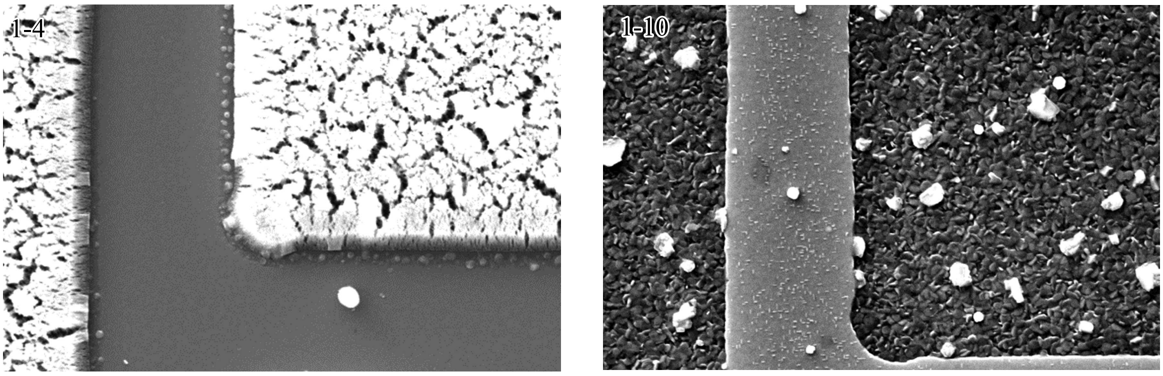


Figure 4. Surface morphology of YBCO in structures L496 and L501. The structures were obtained at the growth temperature of 835°C. Zoom 50′000.

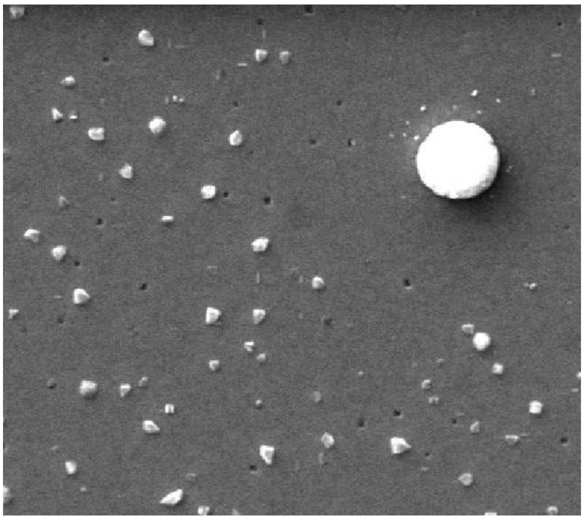


Figure 5. Fragment of structure L479 with defects typical for YBCO epitaxial films upon laser sputtering: one condensation droplet (light, round); precipitates (in large quantities, light, irregular shape); pores (large, dark, irregular).

are faceted particles of irregular shape and with sizes of $\sim 0.5\,\mu$m, pores, and round YBCO condensation drops. The listed types of defects are clearly seen in Fig. 5, which, for example, shows zoomed fragment of a photo of the structure L479.

Specific defects can appear in the TM windows, which are large precipitates with a high density, localized along the TM boundary in a narrow band less than one micrometer wide. Condensation droplets are present on all the structures obtained by us by laser sputtering. These droplets are an artifact of laser sputtering, which results from the material condensation in the gas phase. Droplets have sizes up to $2\,\mu$m, and upon their fall on the substrate, they do not migrate over its surface, but are immediately fixed in the place where they fell, so the gettering properties of the TM do not act on them. The density of condensation droplets is about $2 \cdot 10^5\,\mathrm{cm}^{-2}$, the density of precipitates and pores in wide regions of the YBCO film is much higher — up to $10^8\ \mathrm{cm}^{-2}$.

Photographs of structures L477 (TM „coldCeO_2“) and L476 (TM „islands“) grown at 888°C are shown in Fig. 1. The morphology of YBCO films on these substrates slightly differs both in the presence of defects and in their density: along the mask–film interface, there is a band about $1\,\mu$m wide, free of CuO precipitates, but this effect is absent for pores. The density of precipitates and pores is the same both in wide areas and near the boundary of the TM, and is $10^8\,\mathrm{cm}^{-2}$ and is maximum for the array of structures considered in the paper.

Photos of structures L479 (TM „coldCeO_2“) and L498 (TM „islands“) grown at 866°C are shown in Fig. 2. The film morphology on sample L479 is similar in both wide and local regions to the film morphology on sample L477. The density of precipitates and pores decreased slightly. On sample L498, on bridges $4\,\mu$m wide, the film is free of both CuO precipitates and pores (defect density is less than $10^5\,\mathrm{cm}^{-2}$). We will call the film with such defect density as „very smooth“. In wide regions, the „very smooth“ surface is observed only near the TM boundary; in the rest portion of the wide region the concentration of CuO precipitates is practically unchanged, which is clearly seen in Fig. 6 (structure L498) at a lower zoom. It is possible that the actual growth temperatures during YBCO deposition on structures L479 and L498 were different due to different types of TMs, namely, L479 looks like a film with the growth temperature higher than L498 had (Fig. 2).

When YBCO is deposited on substrates with other options of TM — on samples L491 with TM „coldCeO_2/hotCeO_2“ (Fig. 6) and L492 with TM „coldYBCO“ (photo not shown), obtained at the growth temperature of 866°C, the density of CuO precipitates decreased in local regions. A region free of precipitates

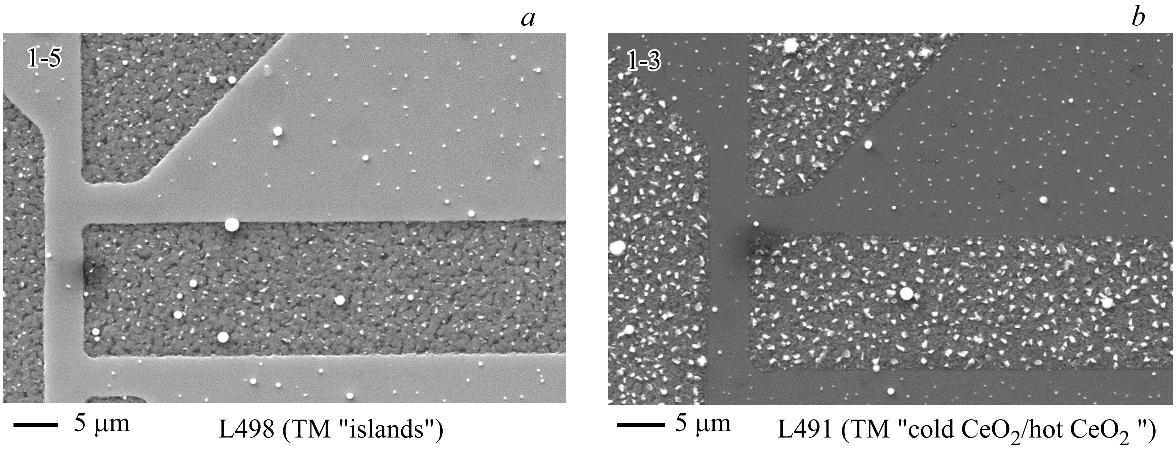


Figure 6. Structures L498 and L491 grown at 866°C. Zoom 15′500.

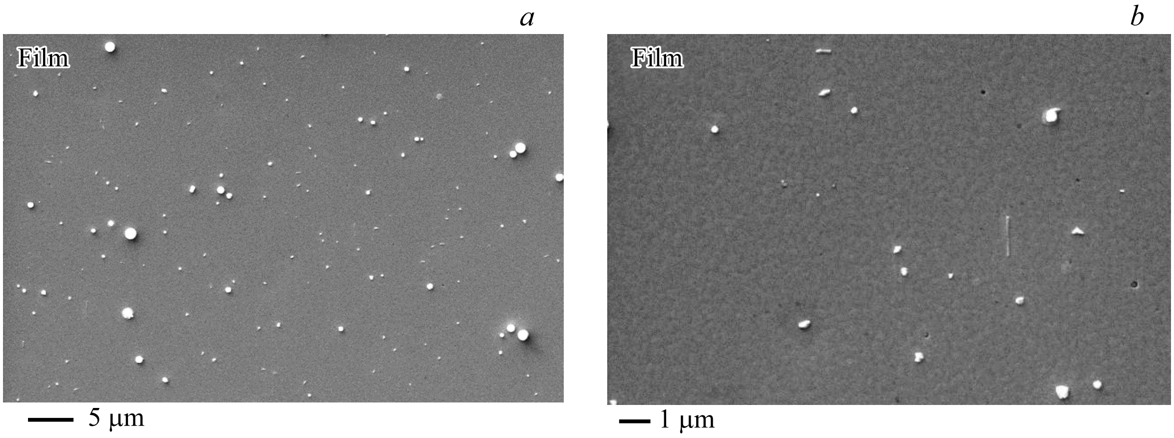


Figure 7. Photos with different zoom (15′500 and 50′000) of a wide region of the YBCO film on the structure L495.

formed along the TM boundary, the region width is greater than at the growth temperature of 888°C. On the same samples a noticeable decreasing of the pores density occurred. Fig. 6 demonstrates the gettering properties of the TM, it clearly shows the formation of a smooth film region along the TM boundary in the presence of precipitates on the rest of the wide region of the YBCO film. This effect — the presence of a smooth film region in narrow regions, and the presence of precipitates in wide regions — was observed in all structures under study at lower growth temperatures. In this case, the defect density in narrow regions was less than $10^5\,cm^{-2}$.

On all structures grown at the growth temperature of 851°C, the presence of a „very smooth“ film was observed in narrow regions. Photos of the structures are shown in Fig. 3: L495 with TM „coldCeO_2“ and L500 with TM „islands“. Fig. 7 shows photos with different zoom of the wide region of the YBCO film on the structure L495, demonstrating the presence of precipitates and pores far from the TM boundary, the precipitate density here is $10^7\,cm^{-2}$. The density of precipitates in narrow regions is less than $10^5\,cm^{-2}$.

With a further growth temperature decreasing to 835°C, the film surface in local regions is also free of defects (density is less than $105\,cm^{-2}$). Photos of the structures are shown in Fig. 4: L496 with TM „coldCeO_2“ and L501 with TM „islands“. Nanometer outgrowths appear on sample L501, oriented in mutually perpendicular directions and, possibly, representing the „*a*-phase“ of YBCO, which was observed by various research groups [7,8]. The observed difference in morphology may be due to the difference in growth temperatures for the L496 and L501 substrates with different masks, as discussed above.

Note that on the photos of structures L495 (Fig. 3) and L496 (Fig. 4) grown on „coldCeO_2“ at temperatures of 851°C and 835°C, respectively, it can be seen that large precipitates with a high density are localized along the TM boundaries, between which a region of a smooth film was formed. In the photo shown (Fig. 3, structure L495), the width of the smooth film region is $2.8\,\mu$m on $4\,\mu$m bridge, and on bridges $10\,\mu$m the size of this region is — $8\,\mu$m (photo not shown). On structures L500 (Fig. 3) and (Fig. 4) L501 with TM „islands“, grown at the same temperatures, this phenomenon is not observed. The study of the nature of this effect and the composition of the observed precipitates is beyond the scope of this work and requires additional studies.

The observed changes in the surface morphology with the growth temperature decreasing from 888 to 835°C can be explained by the fact that at a higher temperature barium re-evaporation from the growth surface occurs, the film composition is enriched with yttrium and copper, and this „excess“ material forms defects — particles of secondary phases. The mask boundary acts as a getter for excess material, resulting in a smooth film formation in local regions. This is clearly seen in the example of structures L498 on substrate with TM „islands“ and L491 on substrate with TM „coldCeO_2/hotCeO_2“ at temperature of 866 °C (Fig. 6). As a result, with the growth temperature decreasing, the YBCO film in local regions becomes practically free of defects (density is less than $10^5\,cm^{-2}$). The defect density in wide regions of the film remains high.

The measurement of electrical properties showed high characteristics on all samples without exception considered in this paper: the critical temperature is about 88 K, the critical current density is more than 3 MA/cm^2 at $T = 77$ K.

4. Conclusion

When YBCO is deposited on the structure with the topology mask by laser sputtering, the extended regions of film with a smooth surface several micrometers wide can be obtained, which, at the same time, have high electrophysical characteristics: a critical temperature of about 88 K, a critical current density of more than 3 MA/cm^2 for $T = 77$ K. Whereas the defect density in the regions of the film far from the boundary of the topology mask remains high and is more than $10^7\,cm^{-2}$.

During laser deposition of YBCO onto substrates with the topology mask in the temperature range of 888−835°C, four types of defects are observed: CuO precipitates; other large precipitates localized along the boundary of the topology mask; pores; condensation droplets. Droplets are the artifact of laser sputtering, their density is about $2 \cdot 10^5\,cm^{-2}$, it is the same both in the narrow windows of the topology mask and on the large regions of the film, and does not depend on the growth temperature. The density of such defects as precipitates and pores differs greatly in the wide regions of the film and in the windows of the topology mask several micrometers wide. On large areas at the growth temperature of 888°C, the density of CuO precipitates and pores is about $10^8\,cm^{-2}$. As the growth temperature decreases, the defect density on large areas, as it should be, decreases ($10^7\,cm^{-2}$ at 851°C).

In the windows of the preliminary topology mask several micrometers wide, the dynamics of changes in the defect density is noticeably different.

− At the growth temperature of 888°C the defect density in narrow regions corresponds to the defect density in wide regions, i.e., $10^8\,cm^{-2}$. But as the growth temperature decreases, starting from 866°C, in the narrow regions of the topology mask the region of a smooth film with a defect density of less than 10^5 per cm^{-2} (less than 1 defect per 1000 μm^{-2}) is formed.

− On substrates with a single-layer topology mask of amorphous cerium oxide, large precipitates with a high density can form, localized along the boundary of the mask. In this case, in the region between the bands of the localized defects a region of smooth film up to 8 μm wide appears with defect density less than $10^5\,cm^{-2}$ (less than 1 defect per 1000 μm^{-2}).

Thus, it is shown that, during YBCO deposition by laser sputtering the preliminary topology mask is an effective getter for defects, which leads to the formation of a rather wide (units of μm) region of smooth film near the boundary of the such mask. This effect is observed for various types of the preliminary topology mask used in this study.

The observed effect makes it possible to form the topology of the YBCO structure with smooth regions of micrometer sizes in places where the critical elements of the superconducting circuit should be located. Therefore, the use of the preliminary topology mask method together with the methods of ion etching or ion implantation can be promising for creating structures with submicron-sized elements.

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Conflict of interest

The authors declare that they have no conflict of interest.

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