STRUCTURAL, TRANSPORT AND MICROWAVE PROPERTIES OF 123/SAPPHIRE FILMS: THICKNESS EFFECT

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ABSTRACT. The effect of thickness and growth conditions on the structure and microwave properties has been investigated for the 123/sapphire films. It has been shown that in the conditions of epitaxial growth the Al atoms do not diffuse from substrate into the film and the films with thickness up to 100nm exhibit the excellent DC properties. The increase of thickness of GdBaCuO films causes the formation of extended line-mesh defects and the increase of the surface resistance (R_s). The low value of surface resistance R_s(75GHz,77K)=20 mOhm has been obtained for the two layer YBaCuO/CdBaCuO/sapphire films.

1. Introduction

High temperature superconducting (HTSC) thin films are currently of interest for microwave applications [1]. The HTSC films for microwave devices would have to be uniform over several inches, smooth, with high density of critical current and low value of microwave losses. Moreover, the film substrates should have a small dielectric constant and low-loss tangent.

In our previous works (see, f.e., [2,3]) we have reported on the epitaxial films of 123 system on sapphire substrates without buffer layer with $j_c(78K) \approx 10^7 A/sm^2$, and also the films on Al_2O_3 substrates with 100 mm diameter, which have been fabricated by the laser deposition technique. However, such high transport properties have been obtained for the films of thickness less 100 nm, i.e. less the penetration depth λ [4]. Smallest values of the surface resistance of HTSC films have been obtained for such thicknesses D which are comparable with λ, D $\approx$ 300 nm.

In this report the effect of the thickness and growth conditions of the films on the crystalline structure, transport and microwave properties has been investigated. Two step deposition allowed us to obtain the relatively low surface resistance of the films, R_s(78K,75GHz) $\approx$ 20 mOhm.

2. Experimental

As it has been described in details in our previous reports [3,5], for the films deposition it have been used the irradiation of a pulse laser (1.06μm) with energy density 8 J/sm^2 and the stoichiometric targets of YBaCuO and GdBaCuO systems. The pressure of ambient oxygen at the deposition was about 0.5 Torr. As the substrates it has been used the sapphire (1012).

In order to better verify the quality of the films the following experimental techniques and equipment were used. The oriental properties [6], crystalline structure and morphology of films were tested by X-ray, Raman spectroscopy and SEM.

The chemical and phase composition of films were tested by differential dissolution (DD) method. This is a new method for characterization of multiphase inorganic materials (chemical method of phase analysis) [7,8]. This method can be shortly described as following.

The film sample under study is placed in reactor, with the time-variated concentration solvent flowing over the film. The solution obtained goes to the detector-analyzer (ICP). Time-variated masses of dissolved elements (except the oxygen) are determined. The regime of dissolution is fixed up such a way that the phases consisting the sample are dissolved differentially (one after another) due to irregularity of their chemical potentials of dissolution. The information obtained is used for stoichiograms construction (set of time- variated molar relations of elements) to interpret the composition and the relative amount of each phase. In this work DD method was used, in particular, to study the interaction character between materials of film and substrate [8].

The transport and superconducting properties of films were investigated by the standard four probe technique and the measurement of DC current-voltage characteristics. For these measurements the film microbridges and meander structures have been patterned by lithography or scribing with acute diamond tool. The Ag or Au contacts to the film samples have been fabricated by the laser deposition technique. The DC critical current of samples was determined by criteria 1μV.

The surface resistance R_s of HTSC films has been measured by the technique [9] of replacing one end wall of a copper cylindrical cavity Ø6.7mm·2.75mm which resonates at frequency of 75.2 GHz in H_{011} circular mode.

3. Results and discussion

In our previous report [2,3] it has been shown that the growth conditions and, correspondently, the quality of laser deposed films depend on a number of technological parameters such as the substrate temperature, characteristics of the laser beam, target-substrate distance, oxygen pressure and the crystalline structure and surface orientation of the substrate materials, too.

Optimal values of the parameters are connected in a such a way that the exchange of one of them can be compensated by correction of other ones. So the exchange of film properties can be obtained by the variation of any of these parameters.

In this work we used the variation of substrate temperature (T_S) to change the film quality and to highlight the correlation of structure, chemical composition and superconducting properties of films.

The kinetic dependences of the dissolution by DD method for two YBaCuO films fabricated at the different T_S are shown on Fig.1. As an example, consider the process of dissolution for the sample deposited at T_S=800°C (Fig.1b). It is seen the impurity phases localized near to the surface layers of the film are dissolved at first. It is also seen that the large content of aluminum atoms is registered that is the consequence of aluminum diffusion from substrate. The following peak on the plot characterizes the dissolution of 123 phase and the impurity phases dispersed in one. And, it is the last, the dissolution of impurity phases localized in the film-substrate region is observed.

The picture of the dissolution of film sample deposited at T_S=780°C (Fig.1a) is distinguished by the absence of aluminum in the surface layer and the smaller concentration of localized and dispersed impurity phases.

The kinetic dependences of dissolution of film samples deposited at other T_S values are like that on Fig.1b.

The orientation characteristics, phase composition and superconducting properties of the films of 80 nm thickness deposited at different T_S are shown in the Table 1. It is seen that the sample deposited at T_S=780°C has the best oriental properties, highest value of critical current density and contains the smallest amount of the impurity phases. In addition, as it has been shown above the not aluminum atoms are registered in the surface film layer.

The next conclusion may be drawn from these results. In the case of optimal growth conditions the epitaxial layers with perfect structure are formed at the initial stage of the film growth. These increased above 100 nm, the structural defects are accumulated in the film in such a way that the influence of substrate orientation is lost and the external layers of thick films have the polycrystalline structure.

One of the reasons of such behavior can be the variation of growth conditions for top film layers at the increasing film thickness. Actually, if the first film layers are deposited on sapphire surface that is the heteroepitaxy occur but the following layers grow as homoepitaxial. So, the optimal deposition conditions must be differed for initial and following film layers.

The investigation of the thickness dependence on surface resistance in this work has been made for the GdBaCuO films. Using the GdBaCuO system allowed us to obtain the thick epitaxial films. The orientation characteristics of top and near substrate-film interface layers of GdBaCuO and YBaCuO thick films to be compared are shown in the Table 2. This result was obtained by Raman spectroscopy technique described f.e. in Ref.[2].

Table 2. The orientation properties of the surface (top) layers and the film layers near the substrate-film interface (bottom layer) of YBaCuO and GdBaCuO films with thickness of 300 nm. Results of Raman spectroscopy investigations.

Material of film	Top layer	Bottom layer
YBaCuO	pollycrystalline	epitaxial
GdBaCuO	epitaxial	epitaxial

The GdBaCuO films with thickness up to 100 nm exhibited the exellent DC properties. In particular, the values of critical current density ($j_c(77K) \sim 10^6-10^7$ A/cm^2) and the transition temperature (T_c=90-91K) were reproduced for the microbridges with the the width 1-10 μm and the length up to 10 cm indicating the high quality of films. But the measurement values of surface resistance were approximately 70 mOhm.

However, at the increasing film thickness the superconducting properties are changed though the orientation properties were high. For the films with thickness about 200 nm the effective value of the critical current density dropped about two times but the surface resistance value was smaller then for the 80 nm thickness films (see Table 3). At the further increasing film thickness to 300 nm the abrupt drop of the critical current density and the increasing of the surface resistance had been observed.

The reason of such degradation of superconducting and microwave properties was the formation of the extended defects in the films. This were the low visible in optical microscope thin lines forming the regularly shaped mesh. The preferred directions of lines correlated with the orientation of a,b-axes of film. The average size of the regions between the mesh lines was up to 10-40 μm. It should be noted that the line defects observed in optical microscope are not registered by SEM.

To visualize the film structure with line defects the films were heated by pulse laser irradiation with the energy density 0.5 J/cm^2. The photography of the surface of thick epitaxial film after irradiation treatment is shown on Fig. 3a. It is seen that the film cracking along the directions of line defects occurs. Unexpectedly, such films after irradiation treatment conserve the superconducting properties but the value of critical current density drops from 10^4 to $10^2 A/cm^2$. It should be noted that at the analogously treatment the cracking thick YBaCuO films take place, too. But the directions of the cracks have the casual character (see the Fig.3b).

The formation of line defects have been registered after the standard post deposition cooling in the oxygen with the pressure of 1 atm at a cool rate of 10 $^{\circ}$C/min. This procedure is used in order to optimize the oxygen content in the film. At the decreasing of cool rate up to 0.5°C/min the formation of defects take place, too. This effect disappeared only in the case when the film samples were cooled at the low oxygen pressure of about 1 Torr. The defects appeared, too at the repeat heating and cooling of the samples at the oxygen pressure of 1 atm.

From this result it may be concluded the effect of line defect formation is bound up with film deformations at the ortho-tetra phase transition which occurs for the oxide superconductors of 123 systems. In our work [2,3] it has been found that the structural mismatching of sapphire and oxide superconductors cause the deformation of film and, correspondingly, changing their structural and superconducting properties. It may be assumed that the increasing film thickness increases the magnitude of deformations at the phase transition and, as result the formation of defects takes place. In the case of YBaCuO films with the polycrystall top layer the visible defects are no formed, because the small size of grains and the large quantity of intergrain boundaries reduce the effect of deformations.

To depress this effect in the thick epitaxial films we used the two layers deposition. The first layer of 60 nm thickness was deposited from GdBaCuO target, and then the following thick layer was deposited from YBaCuO target. Both layers were deposited at substrate temperature of 780°C.

The concept of such two layer deposition consisted of using the difference of the growth rate of yttrium and gadolinium based oxide superconductors. The anisotropy of the growth rates along c-axis and in a,b-plane is smaller for yttrium system then for gadolinium one. So, the size of the monocrystalline regions in the yttrium based films is assumed be smaller then for the gadolinium based films.

Actually, using the two layer deposition allows us to fabricate the films without visible defects and with relatively low surface resistance. It seems likely that at two layer deposition the amount of boundaries between monocrystalline regions are increased in the yttrium based film layer and these defects accumulate the film deformations. But these changes of the film structure should be lowered the effective value of critical current density, that is observed in experiments.

Table 3. The properties of GdBaCuO films versus the film thickness

Film thickness, nm	Critical current density (77K), A/cm^2	Surface resistance (75.2 GHz, 77K), mOhm
80	$8*10^6$	70
200	$3*10^6$	60
300-400 (with defects)	10^4_10^5	$\geqslant$300
300 (two layer deposition)	$6*10^6$	20

In Table 3 the transport and microwave characteristics of HTSC films with different thickness, including two layer samples are shown. It is seen the best result has been obtained for the two layer deposited film.

However, it is not correctly to conclude from here that the two layer deposition is the only way to obtain the good microwave properties for the films on sapphire substrates. We guess that by optimization of growth conditions the films with low surface resistance can be fabricated using the only one target. The our further investigations will be continued in this direction.

4. Conclusions

We have shown that in the 123/sapphire films of thickness more than 100 nm it can occur the formations of extended line defects with regular structure at ortho-tetra phase transition. The films without such defects and with the low value of the surface resistance have been obtained by using the technique of two layer deposition.

This work was supported in part by Committee of HTSC problems (grant 91091) and in part by International Science Foundation.

5. References

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