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Superconducting properties of YBaCuO thin films obtained by thermocycling

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YBaCuO films fabricated by laser ablation method were subjected to a thermocycling. Our investigations showed that the degradation process of the current transport properties of films is accompanied by the appearance of a maximum on the curve of the critical current density, depending on the number of thermocycles. This effect manifests itself most strongly in the films with degraded superconducting properties. The critical temperature and the transition width vary only slightly in this case.

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1. Introduction

The use of HTSC films for the manufacturing of sensors, SQUIDs, flux transformers, and other devices [1–5] inevitably raises the question of how the sensitive elements from the HTSC films will behave upon multiple cooling to the boiling temperature of nitrogen and heating to the indoor temperature, i.e., during thermocycling. It is important to test the films under conditions close to real at applied point of view. At present, there are no sufficiently complete investigations of this matter in the current literature [6–8]. We have therefore conducted a study of the superconducting properties of YBaCuO films at thermocycling. The results of this study are presented in this paper.

2. Film manufacturing procedure. Thermocycling

The YBaCuO thin films have been deposited by laser ablation method [9]. As a substrate we used single-crystal plates of $SrTiO_3$ (100), $LaAlO_3$ (100) and sapphire (100). Polycrystalline pellets of YBaCuO 1 cm in diameter, 0.5 cm thick, and 4.4–4.6 g/cm^3 in density were used as a target. We used a pulsed Nd:YAG laser (wavelength $\lambda = 1.06\,\mu$m, pulse length $\tau = 20$ ns, repetition race $\nu = 12$ Hz). The substrate temperature was adjusted at temperature in the range 810–840 °C, and the oxygen partial pressure in the vacuum chamber was about 0.1–0.6 mbar during the deposition. The power density of laser radiation on the target surface varied from $3\cdot10^8$ W/cm^2 to $8\cdot10^8$ W/cm^2. The deposition rate was about 50 nm/min, and the cooling rate after the deposition was about 25 °C/min. Superconducting properties of films obtained by laser ablation method are very sensitive to a variation of the deposition parameters. A small variation of deposition parameters, even one of them, leads to a considerable change in the critical temperature T_c, transition width ΔT, critical current density J_c, and film structure.

A thermocycling procedure consists of cooling a test sample to the nitrogen boiling temperature and heating to room temperature in the air. The cooling rate was about 25 °C/s, and the heating rate was about 40 °C/s. The studied films were made in the shape of long strips of width about $10\,\mu$m and length about $72\,\mu$m by means of focused ultraviolet laser beam. The critical current was measured by a four-probe method. The 1-μV criterion was used for the critical current. Critical current measurements were carried out at 77 K. The samples with different initial superconducting properties [T_{c0}, ΔT, J_{c0}(77 K)] were used.

3. Experimental results

In spite of the expected monotonic degradation of superconducting film properties during thermocycling, we have obtained a rather surprising result. The typical behavior of the films can be described as follows: the values of T_c and ΔT remain virtually the same (for some samples deposited on sapphire up to 1400 thermocycles), but the dependence of the criti-

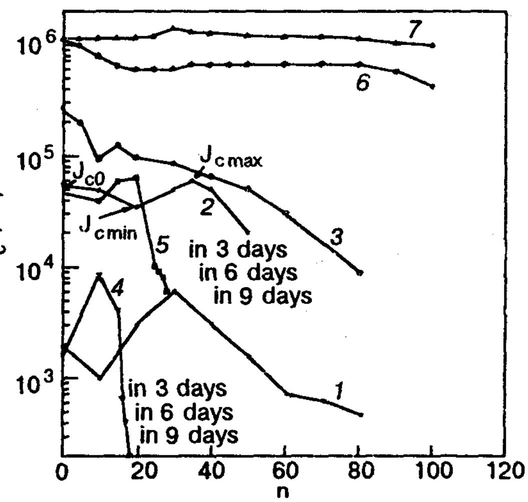

Fig. 1. J_c vs the number of thermocycles n for the samples 1–7 on the $SrTiO_3$ substrates. The last three points for the samples 4 and 5 correspond to measurements in 3, 6, and 9 days, respectively.

cal current density J_c on the number of thermocycles n has a maximum; the values this maximum may sometimes exceed the initial value J_{c0}. This effect differs numerically for different film samples.

The J_c dependence on the thermocycle number n for samples 1–7 deposited on the $SrTiO_3$ substrates is shown in Fig. 1, and the same dependence for samples 8 and 9 is shown in Fig. 2. Table 1 gives the values of the initial critical current density J_{c0} for samples 1–9, the ratios J_{cmax}/J_{c0} and J_{cmax}/J_{cmin}, calculated from the experimental data, where J_{cmax} and J_{cmin} are the values of the critical current density at the maximum and minimum of the $J_c(n)$ curve,

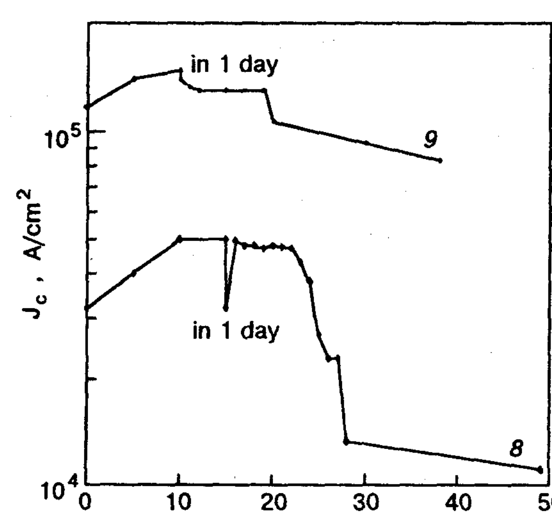

Fig. 2. J_c vs the number of thermocycles n for the samples 8–9 on the $SrTiO_3$ substrates.

respectively, n_{min}, corresponding to J_{cmin}, is shown to the left of n_{max}, corresponding to J_{cmax}. The points corresponding to J_{c0}, J_{cmax}, and J_{cmin} for sample 1 are indicated in Fig. 1.

Table 1

Sample	J_{c0}, A/cm^2	J_{cmax}/J_{c0}	n_{max}	J_{cmax}/J_{cmin}	n_{min}
1	$1.6\cdot10^3$	2.5	30	3.6	10
2	$5.5\cdot10^4$	0.7	35	1.8	20
3	$2.69\cdot10^5$	0.48	15	1.1	10
4	$1.55\cdot10^3$	4.7	10	4.7	0
5	$4.7\cdot10^4$	0.9	20	1.8	10
6	$1.11\cdot10^6$	0.6	35	1.1	15
7	$1.16\cdot10^6$	1.2	30	1.2	10
8	$3.19\cdot10^4$	1.7	10	1.7	0
9	$1.17\cdot10^5$	1.3	10	1.3	0

A comparison of the data for samples 1–3 shows that the ratios J_{cmax}/J_{c0} and J_{cmax}/J_{cmin} can be lowered by increasing J_{c0} from one sample to the other. In this case T_{c0} is equal to 88.0, 89.2, and 89.0 K and ΔT is equal to 3.6, 4.8, 4.0 K for samples 1–3, respectively (see Table 2).

Table 2

Sample	T_{c0}	ΔT_0	$T_c(n_{max})$	$\Delta T(n_{max})$	$T_c(n')$	$\Delta T(n')$
	K					
1	88.0	3.6	88.4	4.0	87.6 (80)	5.2 (80)
2	89.2	4.8	88.8	4.2	89.4 (50)	4.4 (50)
3	89.0	4.0	89.0	4.0	89.0 (80)	4.2 (80)
4	89.4	3.4	90.0	3.6	89.4 (9 days)	3.6 (9 days)
5	89.2	2.0	89.0	2.2	89.0 (9 days)	2.2 (9 days)
6	91.2	2.4	91.2	2.4	91.0 (120)	2.2 (120)
7	91.2	1.6	90.8	1.8	90.6 (110)	2.0 (110)
8	89.8	2.4	91.2	2.2	90.2 (50)	2.4 (50)
9	90.2	2.2	90.4	2.2	90.6 (38)	2.0 (38)

In other words, the initial critical temperature and the initial transition width do not exert much influence on the observed dependence $J_c(n)$, but the ratios J_{cmax}/J_{c0} and J_{cmax}/J_{cmin} are sensitive to the initial value of J_{c0}. It is hardly possible to establish here some kind of correlation n_{max} or n_{min} with the initial values J_{c0}, T_{c0}, and ΔT_0. As can be seen from Fig. 1, for all three samples (1–3), after going through the maximum J_c decreases qradually with increasing n.

Samples 4 and 5 were returned to normal conditions on the third, sixth, and ninth day (Fig. 1). It is seen that the degradation process of the current transport properties of these films continue. Samples 8 and 9 were also returned to normal conditions after 1 day, but, in contrast to samples 4 and 5, after reaching J_{cmax} (see Fig. 2). In this case the relaxation of J_c to the initial value occurred in sample 8; in sample 9 only a tendency to this relaxation is appreciable.

The effect of critical current growth during thermocycling is faintly expressed in samples 6 and 7 (Fig. 1), which possess high values of J_{c0} ($1.11 \cdot 10^6$ and $1.16 \cdot 10^6$ A/cm^2 for samples 6 and 7, respectively). The samples have no clear maximum. Both samples degrade insignificantly even after 100 thermocycles.

The values of T_c and ΔT were measured simultaneously with the measurement of J_c, but in Table 2 only the initial values of T_{c0} and ΔT_0, the values of $T_c(n_{max})$ and $\Delta T(n_{max})$, corresponding to n_{max}, and also the values of $T_c(n')$ and $\Delta T(n')$, where n' is the total number of thermocycles for the given sample, are presented. The results which we obtained confirm that the effect of the increase in the critical current density essentially does not correlated with the behavior of T_c and ΔT for all samples. In other words, the degradation of the current transport properties of

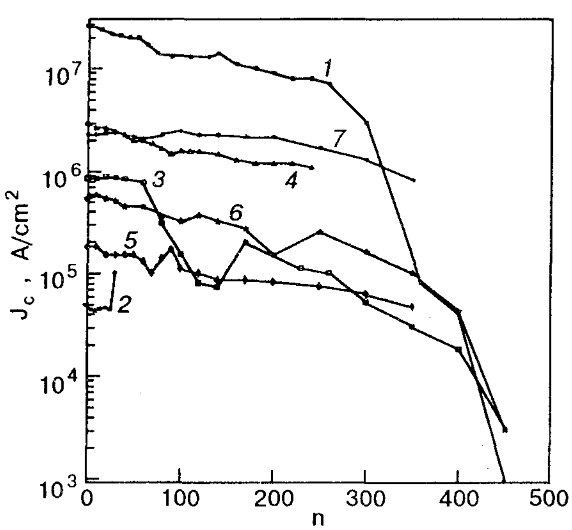

Fig. 3. J_c vs the number of thermocycles n for samples 1–7 on the $LaAlO_3$ substrates.

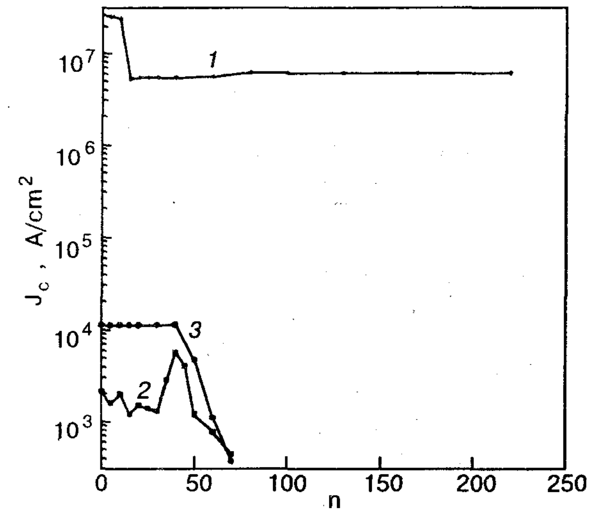

Fig. 4. J_c vs the number of thermocycles n for samples 1–3 on the sapphire substrates.

films is not accompanied by a simultaneous decrease in the critical temperature and an increase in the transition width.

A similar behavior of J_c vs n was observed in the films deposited on the $LaAlO_3$ substrates (Fig. 3) and on the sapphire substrates (Fig. 4). There are, however, some differences. For example, the largest value of J_{cmax}/J_{cmin}, equal to 4.77 (sample 2 in Fig. 4), was obtained for a film deposited on sapphire with the initial value of $J_{c0} = 2.2 \cdot 10^3$ A/cm^2. Further, the film deposited on sapphire with $J_{c0} = 2.6 \cdot 10^7$ A/cm^2 (sample 1 in Fig. 4) showed the highest stability to thermocycling: after 1 400 thermocycles the critical temperature remained the same, $T_{c0} = 91.6$ K and the transition width also was nearly the same: $\Delta T_0 = 2.0$ K, $\Delta T_{1400} = 2.2$ K, but the critical current density changed from $2.6 \cdot 10^7$ to $4.1 \cdot 10^6$ A/cm^2.

4. Discussion

The results obtained by us show that the films grown by laser ablation method are not single-crystalline in all the film volume, and that their macroctructure consists of crystal grains divided by boundaries between them. Our investigations of the films with the scanning microscope JEOL JSM-840 showed that the oriented single-crystal grains have the following characteristic dimensions: 0.5–1.5 μm in length and 0.1–0.2 μm in width for samples with the low initial values of J_{c0}. A slight dependence of the critical temperature T_c and the transition width ΔT on the number of thermocycles indicates that these parameters generally define the bulk superconducting properties of the grains. As for the current transport properties of films, they are determined mainly by the integrain boundaries.

It is known that the properties of intergrain boundaries are defined by many parameters [11–18], but we choose only one of them which most probably characterizes the processes taking place during thermocycling. They are: 1) the intergrain disorientation angle θ in the *ab* plane [17], 2) an oxygen stoichiometry in the intergrain fields and a diffusion of atoms and molecules from the environmental area, and 3) a characteristic of the superconducting weak link of the grain contact.

The critical current density at the intergrain boundary, J_c^{GB}, depends considerably on the angle θ of the grain disorientation [10–13] and J_c^{GB} is quickly lowered with an increase in θ. For example, an exponential dependence of J_c^{GB} on the angle θ is presented in Ref. 10:

$$J_c^{GB} = J_c^G \exp(-\theta/\theta_0) \ , \qquad (1)$$

where J_c^G is the critical current density in the grain, and $\theta_0 = 5°$. Other approximations are used in Refs. 11 and 12, but all of them correspond to a rapid lowering of J_c^{GB} with an increase in θ. On the other hand, it is known that most of the intergrain boundary is comprised of the Josephson junctions, for example, of the species *SNS* [13,16–18], and in the case $d_N \geq \xi_N$ (d_N is the intergrain layer thickness in the normal state, and ξ_N is the coherence length in the material of this layer) the critical current of this transition is described by the equation [19]

$$J_c \sim \frac{\Delta^2}{\xi_N} \zeta \exp(-d_N/\xi_N) \ , \qquad (2)$$

where Δ is the order parameter in the grain, ζ is a factor which takes into account the decrease of the order parameter in the grain because of the proximity effect. Although Eqs. (1) and (2) were obtained by different methods, it is none the less quite obvious that there is a connection between them, since they describe the same current through the intergrain boundary — the Josephson junction. If this is correct, we may assume that d_N and θ are functionally connected with each other: $d_N = f(\theta)$ and d_N is probably proportional to θ, $d_N \sim \theta$, since $d_N \geq \xi_N$. In this case the disorientation angle θ, defined, in general, by the technological parameters of the film growth, sets the intergrain layer thickness d_N. The observed increase in J_c during thermocycling may therefore be connected with the lowering of the disorientation angle θ and, consequently, with the decrease in d_N. What is the physical cause of the lowering of θ during thermocycling? There is no exact answer to this question so far. We can make only some assumptions. It is known that deformation of a high-temperature superconductor lattice leads to the deterioration of the superconducting properties of the material and also to the total destruction of these properties [20]. The value of elastic deformation increases with an increase in θ. Therefore, it is possible that thermocycling initially reduces the extent of elastic deformation near the boundary layers, which leads to a decrease in θ and to an increase in the critical current density. The boundary properties become degraded as a result of the subsequent thermocycling because of the diffusion of atoms and molecules from environmental area and the accumulation, in particular, of H_2O molecules in the intergrain fields. We assume that the Josephson intergrain junction undergoes many changes of the following kind: $SNS \rightarrow SN'S \rightarrow SN'IN'S$, where I is an insulator, and $d_N < d_{N'}$.

It is evident that the intergrain disorientation angle can be decreasing by increasing $J_{c0} > 10^5$ A/cm^2; therefore, the thermocycling influences the critical current slightly and the effect of critical current growth can be evaluated to some extent. It is possible that the film on all three substrates is a single-crystalline film at $J_{c0} > 10^6$ A/cm^2; therefore, the effect disappears and the films are very stable to the thermocycling in this case.

5. Conclusion

The results which we obtained anable us to make the following conclusions: 1) The degradation process of the current transport properties of YBaCuO films is accompanied by the appearance of a maximum on the curve of $J_c(n)$, 2) this effect becomes more pronounced as the initial superconducting properties are degraded, and 3) the films with high initial values of J_c have very high stability to the thermocycling. It is obvious that a more complete understanding of the critical current increase in YBaCuO films during thermocycling required further studies.

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