Surface Impedance of $YBa_2Cu_3O_{7-\delta}$ Films Grown on MgO Substrate as a Function of Film Thickness

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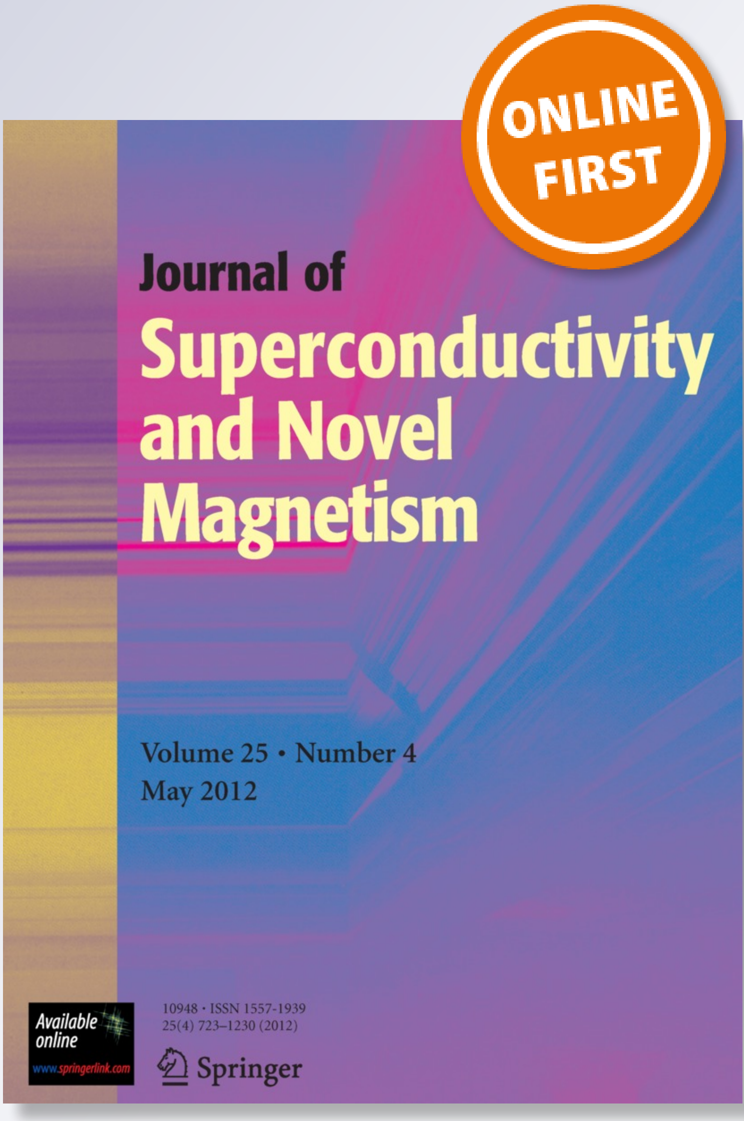

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ORIGINAL PAPER

Surface Impedance of $YBa_2Cu_3O_{7-\delta}$ Films Grown on MgO Substrate as a Function of Film Thickness

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Abstract The surface impedance characteristics of epitaxial $YBa_2Cu_3O_{7-\delta}$ films of thickness $d_f = 75, 150, 300, 600$ nm, produced by magnetron thermal co-evaporation onto single crystal MgO substrates was studied using measurement technique based on Ka-band whispering gallery mode (WGM) dielectric resonator (DR) fabricated from single crystal sapphire. Characterization of the unpatterned films was carried out in temperature interval from 20 K to 90 K. It was shown that the effective surface resistance approaches the minimum value for $d_f > 300$ nm. At the same time, intrinsic impedance properties are practically independent on d_f in the studied interval of d_f values. The temperature dependence of London penetration depth was estimated experimentally and approximated with the model expressions. Effect of reducing the surface resistance of approximately two times at low temperatures one year later after their manufacture was registered for all films(except the film of 75 nm thickness). The effect may be explained by changes of the film parameters in time after the film light overdoping.

Keywords YbaCO thin films · Surface impedance · Quasioptical resonator

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1 Introduction

High-temperature superconductors (HTS) on the basis of cuprate materials are used in microwave (MW) technique in a form of films grown on low-loss single crystal dielectric Al_2O_3, $LaAlO_2$, MgO, and some other substrates (see, e.g., [1]). The high quality films are also used for physical study of HTS using microwave impedance approaches (see, e.g., [2–4]). MW impedance studies of HTS films have two peculiarities determined by two external factors. The first factor is defined by finite thickness of the film, d_f, therefore in the general case the ratio between d_f and London penetration depth $\lambda_L(T)$ in superconductor has to be taken into account. The second factor is related with specific structures used for measure of surface impedance, for example, structures based on coplanar transmission lines [5]. Here, the film is subjected to technical processing, which can result in degradation of initial superconducting material [6].

A number of papers [6–10] is devoted to a problem of impedance properties of HTS films dependence on film thickness d_f, however, previously were studied films sputtered onto substrates such as Al_2O_3 and $LaAlO_2$. At the same time for practical applications, MgO substrates are often used. The dielectric permittivity of MgO crystal is near to permittivity of Al_2O_3 one, however, isotropy of the crystal and possibility of deposition of the HTS film without buffer layer are the advantages of MgO [1, 11]. In addition, the thermal expansion coefficient of MgO ($\alpha = (13-14) \cdot 10^{-6}$/K) 1.5–2 times larger than corresponding coefficient values of Al_2O_3 or $LaAlO_2$ [1]. This can be useful to design the MW devices with electromagnetic structures on the basis of HTS and normal conductors. In this respect, the optimization of film thickness for design of MW devices such as microwave power limiter [12–14] is necessary.

Thus, the main task of our work was study of surface impedance properties of unpatterned $YBa_2Cu_3O_{7-\delta}$ films

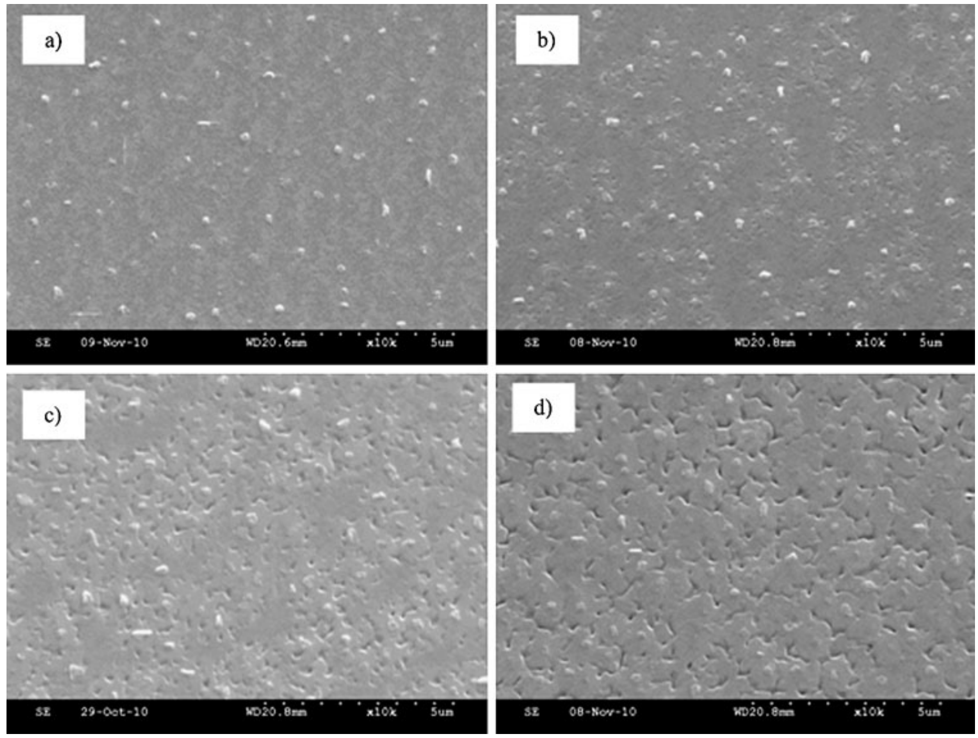

Fig. 1 The SEM images for $YBa_2Cu_3O_{7-\delta}$ thin films (thermal co-evaporated on MgO substrate) of different thicknesses, d_f (nm): (**a**) 75, (**b**) 150, (**c**) 300, (**d**) 600

thermal co-evaporated onto MgO substrates (YBaCuO/MgO structures). Impedance dependence on thickness d_f in a wide range of d_f values as well as film changes of properties in time are studied. From this point of view, we used high-sensitive surface impedance measure techniques, which allow us to measure precisely properties of unpatterned films.

2 Experimental Details and Basic Mathematical Correlations

We experimentally studied epitaxial $YBa_2Cu_3O_{7-\delta}$ films with thickness $d_f = 75, 150, 300, 600$ nm, produced by magnetron thermal co-evaporation onto single crystal MgO substrates of 0.5 mm thickness and 20×20 mm^2 area. Earlier this technology allowed to produce the best quality in situ $YBa_2Cu_3O_{7-\delta}$ thin films [11], although at present researches state that all deposition technique are able to produce high quality thin films with excellent microwave properties [6]. Main properties of the studied films are listed in Table 1. The values of T_c and J_c were measured inductively.

The images of the film surface, obtained by scanning electron microscope (SEM), are shown in Figs. 1a–1d.

The films investigated in this study were optimized for low surface resistance and had excess of yttrium and copper. This non-stoichiometric composition leads to a characteristic morphology, in which the surplus of copper appears as copperoxide particles on the surface (white precipitates with diameter of several 100 nm). In contrast, the yttrium excess

Table 1 Main properties of the studied films

Thickness (nm)	T_c (K)	J_c (MA/cm^2)
75	85.8	3.0
150	86.5	3.6
300	87.1	3.2
600	87.6	2.9

leads to formation of pores, which can be seen most clearly for films with thickness of 300 nm and above.

For MW impedance characterization of large-area unpatterned HTS films, earlier the authors developed the measurement technique based on whispering gallery mode dielectric resonator (WGM DR) made of single crystal sapphire [15]. In a given work, WGM DR was used in a shape of hemisphere (Fig. 2) [16, 17]. The values of Q-factor and frequency shift of the resonator were measured in Ka-band using network analyzer HP8510C.

It is known that dependence of YBaCuO/MgO impedance properties in low temperature region ($T < 20$ K) can show peculiarities related with peculiarities of complex permittivity $\varepsilon(T)$ in MgO [18]. We did not have the opportunity to study $\varepsilon(T)$ of MgO in this interval of T and we were interested in more "practical" temperature interval ($T > 40$ K), therefore, we carried out measurements in a range from 20 K to 90 K. Because of removal of the WGM two-field degeneration problem [15], the authors analyzed resonance lines using a special computer program [19].

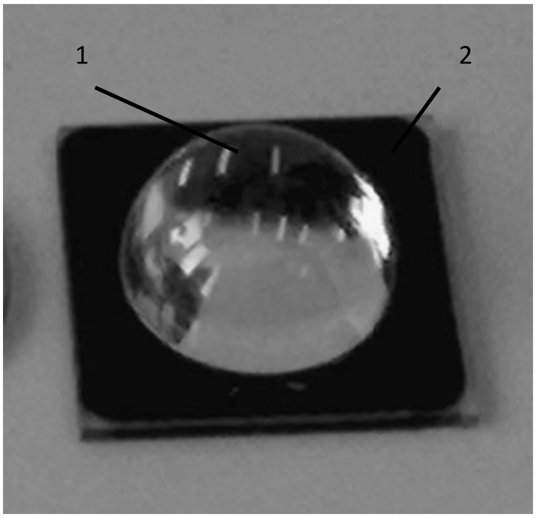

Fig. 2 WGM resonator in a form of sapphire hemisphere (*1*) with HTS film endplate (*2*); optical *c*-axis is perpendicular to conducting endplate

In samples under study, as a rule, the film thickness d_f of the order of London penetration depth λ_L, therefore, the measured surface impedance $Z_s^{\text{eff}} = R_s^{\text{eff}} + iX_s^{\text{eff}}$ depends on properties of both HTS film and substrate. Here, an approach based on the transformation rule of impedance can be applied [1]

$$Z_s^{\text{eff}}(d_f) = Z_s \frac{Z_s \tanh(kd_f) + Z_d}{Z_s + Z_d \tanh(kd_f)} \tag{1}$$

where $k = 1/\lambda_L$ is the magnitude of wave number, $Z_s = R_s + iX_s$ is a surface impedance of the bulk HTS (intrinsic Z_s) Z_d is the impedance of dielectric substrate, k, Z_d, Z_s^{eff} are functions of temperature. As a further shown in [20, 21], the film approximation of (1) in a case of small dielectric loss (i.e. loss tangent $\tan(\delta)$ is small) gives the expression

$$Z_s^{\text{eff}}(d_f) \approx Z_s \coth\left(\frac{d_f}{\lambda_L}\right) \tag{2}$$

which enable to obtain correct results in the range $0.01 \leq d_f/\lambda_L \leq 1$.

Effective surface resistance is found from the measured Q-factor value by expression [15, 17]

$$R_s^{\text{eff}} = \frac{Q^{-1} - k\tan(\delta)}{A_S} \tag{3}$$

where k and A_s are coefficients describing interaction of MW fields with the dielectric material of WGM DR and HTS film accordingly.

Experimental finding of surface reactance X_s^{eff} is more difficult because we cannot measure eigen frequency of DR with perfect conductor (see, e.g., [17]).

However, the measurements of the temperature change of resonance frequency $\Delta f(T)$ can be used to find the change of $X_s^{\text{eff}}(T)$

$$\Delta X_s^{\text{eff}} = \frac{2\Delta f(T)}{A_s f_0(T)} \tag{4}$$

If $\lambda_L(T) = \lambda_L(0)$ is known, we can write

$$X_s^{\text{eff}} = X_s^{\text{eff}}(0) + \Delta X_s^{\text{eff}}(T) \tag{5}$$

where X_s^{eff} is calculated using (2) and $X_s^{\text{eff}}(0) = \omega\mu_0\lambda_L(0)$, where $\Delta f(T)$ is the measured frequency shift in respect to the resonance frequency $f(T = 0)$. However, in practice Δf and ΔX_s^{eff} are measured referring to a lowest temperature T_r, which can be obtained in experiment. The difference $\Delta X_s(T_r) = X_s(T_r) - X_s(0)$ can be usually neglected, although it can be estimated by approximation of temperature dependence $\Delta X_s(T)$ taking into account $X_s(0)$.

The obtained values of $R_s^{\text{eff}}(T)$ and $X_s^{\text{eff}}(T)$ allow us to find the $R_s(T)$ and $X_s(T)$ values for (1) or (2), which characterize bulk or intrinsic properties of the film.

For physical studies, the more valuable expressions are

$$\sigma_n = \frac{2\omega\mu_0 X_s R_s}{|Z_s|^4} \tag{6}$$

$$\sigma_2 = \frac{\omega\mu_0(X_s^2 - R_s^2)}{|Z_s|^4} \tag{7}$$

where $|Z_s|^2 = R_s^2 + X_s^2$, which follow from $Z_s = \sqrt{i\omega\mu_0/(\sigma_1 - i\sigma_2)}$. Here, $\sigma_1 = \sigma_n$ is conductivity of quasi-particles, $\sigma_2 = i\sigma_s$ is determined by properties of paired electrons (conductivity of superfluid electron component).

The conductivity

$$\sigma_2 = \frac{1}{\omega\mu_0\lambda_L^2} \tag{8}$$

allows us to find $\lambda_L(T)$.

3 Results and Discussion

Results of temperature dependencies of $R_s^{\text{eff}}(T)$ and $\Delta X_s^{\text{eff}}(T)$ obtained using (3) and (4)–(5) and experimentally measured Q-factor and frequency shift $\Delta f(T)$ of the resonator with HTS films are shown in Figs. 3a, 3b. Here, we used value of London penetration depth at $T = 0$, $\lambda_L(0) = 160$ nm well established in other studies (e.g., see [2]) in calculations. It is also assumed that $\lambda_L(0)$ does not depend on d_f, at least in the range of films thickness studied. Frequency shift $\Delta f(T)$ was measured in respect to frequency f at $T = 20$ K, therefore, curves $\Delta X_s^{\text{eff}}(T)$ begin from $\Delta X_s^{\text{eff}} = 0$ for all films (Fig. 3b).

For practical applications, it is of important to know effective value $Z_s^{\text{eff}}(T)$, especially $R_s^{\text{eff}}(T)$ at $T = 77$ K, because it determines energy loss in HTS-based MW devices.

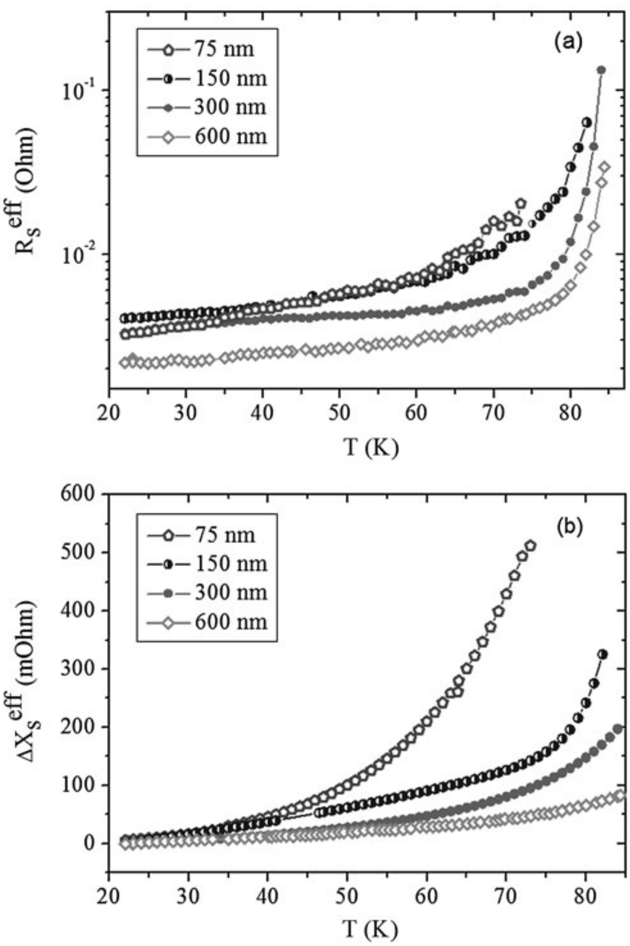

Fig. 3 The temperature dependencies of (**a**) effective resistance (R_s^{eff}) and (**b**) effective surface reactance difference ($\Delta X_s^{\mathrm{eff}}$) of the $YBa_2Cu_3O_{7-\delta}$ thin film thermal co-evaporated onto MgO substrate

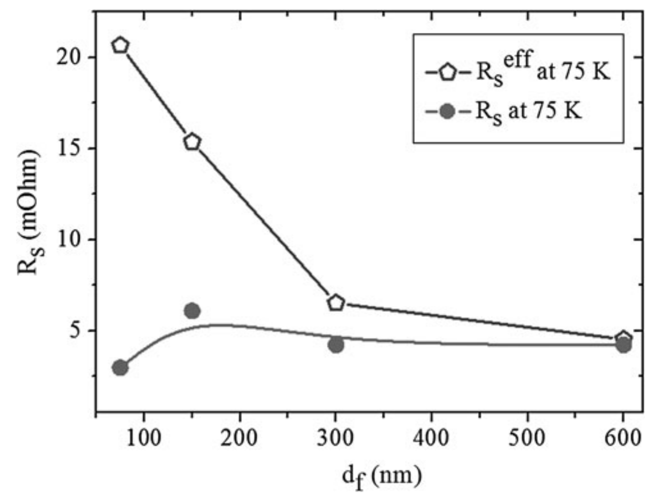

Fig. 4 Effective surface resistance (R_s^{eff}) and intrinsic surface resistance (R_s) of $YBa_2Cu_3O_{7-\delta}$ thin films as functions of film thickness (d_f) at $T = 75$ K

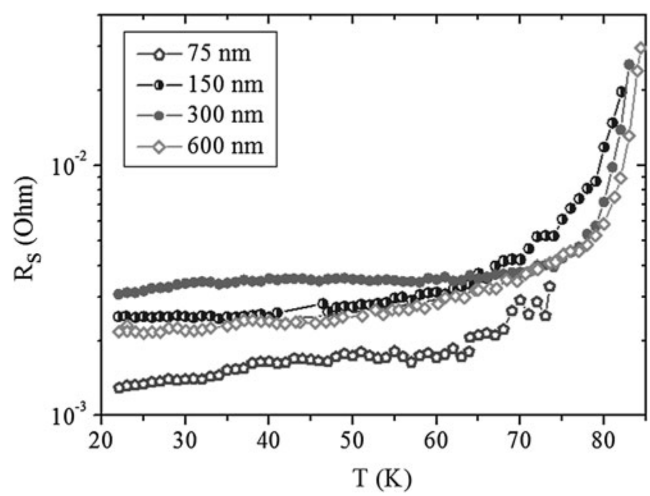

Fig. 5 Intrinsic surface resistance (R_s) of $YBa_2Cu_3O_{7-\delta}$ thin films as a function of temperature T

Taking into account different frequency behavior: for superconductors $R_s \sim f^2$ against $R_s \sim f^{1/2}$ for normal conductors, the values of R_s get close to some critical difference. This indicates on reactance difference ($\Delta X_s^{\mathrm{eff}}$) (Fig. 3b) of the $YBa_2Cu_3O_{7-\delta}$ thin film sputtered onto MgO substrate and on advantages of HTS films utilization up to the frequencies do not exceed a certain frequency f_c, called "critical frequency" [22]. Here, one exception should be noted when HTS films are used in power limiters of microwave signals [12–14].

Measurement results show, that the thickest film ($d_f =$ 600 nm) has the smallest losses at $T = 75$ K, and thinner films ($d_f = 75$ nm and 150 nm) have the largest losses (Figs. 3a, 4). The influence of the substrate is clearly seen in curves $\Delta X_s^{\mathrm{eff}}(T)$ for different d_f (Fig. 3b). In the region $T > 40$ K, the slope of $\Delta X_s^{\mathrm{eff}}(T)$ strongly increases with increasing of d_f. It can be explained by the fact that with increasing of T films become more transparent for MW field. And the same time influence of the substrate becomes stronger at growth of temperature.

Theoretically intrinsic meanings $R_s(T)$ for each d_f has to be the same. Thus general convergence of curves $R_s(T)$ in Fig. 5 is understandable completely. However, significant decrease of $R_s(T)$ for film of $d_f = 75$ nm in respect to other films contradicts the established concepts of deterioration of the films with small thickness. Unfortunately, obtained data are insufficient for an unambiguous interpretation of dependence R_s vs. d_f. On the other hand, it can be stated that if the dependence exists, it is weak.

The obtained values of $X_s(T)$ allow us to estimate dependence $\lambda_L(T)$ in accordance with (3), (5), (7), and (8). Herewith, there is a qualitative difference in the nature of dependence $\lambda_L(T)$ for film of $d_f = 75$ nm from $\lambda_L(T)$ for other films (Fig. 6). These dependences for all thickness val-

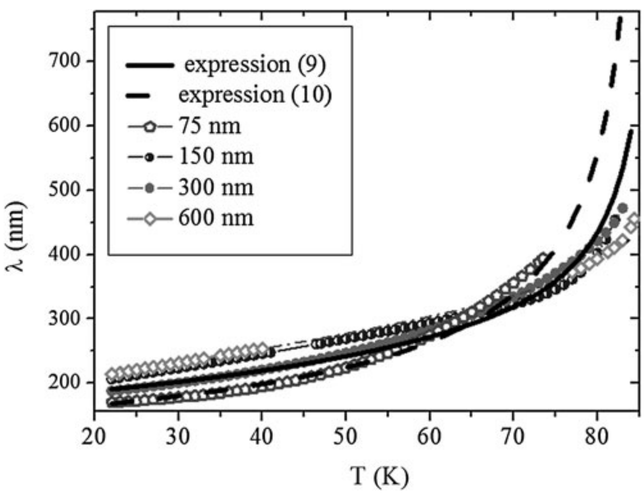

Fig. 6 London penetration depth (λ_L) of $YBa_2Cu_3O_{7-\delta}$ thin films as a function of temperature T. *Solid* and *dashed lines* represent results of approximation obtained using (9) and (10)

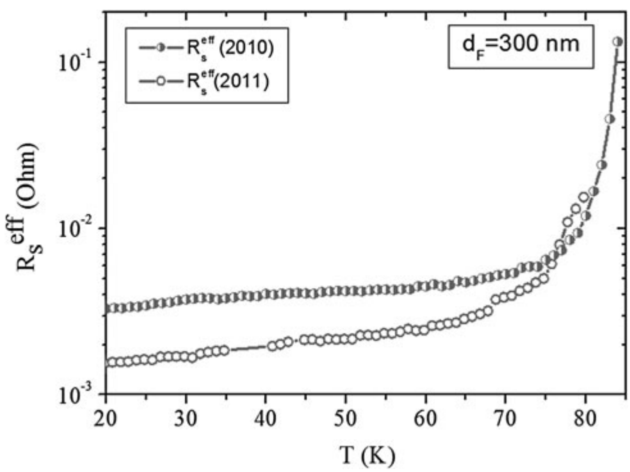

Fig. 7 Comparison of effective surface resistance (R_s^{eff}) data obtained in two measurement cycles for film of $d_f = 300$ nm: 2010 year (*filled symbols*) and about one year later (*empty symbols*) after its manufacturing

ues except 75 nm can be approximated well by the expression

$$\frac{\lambda(T)}{\lambda_L(0)} = 0.165 e^{\frac{0.028}{T}}. \tag{9}$$

The dependence $\lambda_L(T)$ for 75 nm can be approximated well by the known expression

$$\lambda(T) = \lambda_L(0)/\left[1 + (T/T_c)^{\gamma}\right]^{1/2} \tag{10}$$

with $\gamma = 1$ and $\lambda_L(0) = 145$ nm.

Approximately one year later, the $R_s^{\mathrm{eff}}(T)$ were measured again for all the films studied earlier. As example, Fig. 7 shows such a dependence $R_s^{\mathrm{eff}}(T)$ for film of $d_f = 300$ nm. It can be seen that new data at low temperatures are approximately 2 times lower in comparison with $R_s^{\mathrm{eff}}(T)$ measured earlier.

This correlation is the same for all films, except film of $d_f = 75$ nm (Fig. 8). However, after the first measurement the central part of this film was damaged, so, perhaps, here the effect of R_s^{eff} decrease was not registered due to the damage. The found effect of significant improvement of microwave properties of $YBa_2Cu_3O_{7-\delta}$ films in time may be explained by the deliberate overdoping level in the film production process at "THEVA" company. It is noting, meanings of $R_s^{\mathrm{eff}}(T)$ were unchangeable practically at temperatures near 77 K.

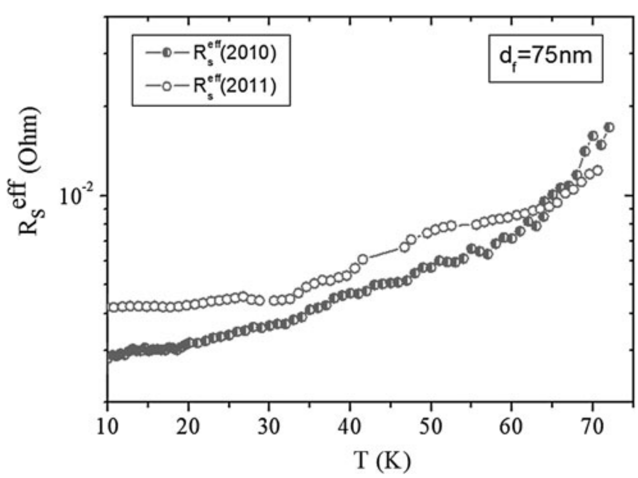

Fig. 8 Comparison of effective surface resistance (R_s^{eff}) data obtained in two measurement cycles for film of $d_f = 75$ nm: 2010 year (*filled symbols*) and about one year later (*empty symbols*) after its manufacturing

4 Conclusion

Thus, in this work, the microwave surface impedance of $YBa_2Cu_3O_{7-\delta}$ films of different thickness ($d_f = 75$–600 nm), thermal co-evaporated onto single crystal MgO substrates of 0.5 mm thickness were studied.

Experimental measurements were performed using Ka-band sapphire resonator in a shape of hemisphere excited with whispering gallery modes. It was experimentally shown that R_s^{eff} decrease tends to the minimum at $d_f > 300$ nm, and intrinsic properties of the films don't depend on d_f in the mentioned range of d_f. London penetration depth can be modeled at $T > 20$ K by the expression (9) for all thickness values except 75 nm. The dependence for the latter is approximated well by the known expression (10) with $\gamma = 1$ and $\lambda_L(0) = 145$ nm.

The effect of considerable (approximately 2 times at low temperatures) reduction of $R_s(T)$ was detected a year later after film synthesis. This effect may be explained by the deliberate overdoping of the films in the synthesis process and their subsequent "aging" process when films become optimally doped.

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