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Microwave Surface Resistance of Plasma-Sprayed YBaCuO Thick Films on Large-Area Metallic Substrates

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The microwave surface resistance of YBaCuO thick films was measured using a demountable copper cavity operated at 3 GHz in the TEO11 mode over a temperature range from 4.2 K to 300 K. The films were deposited onto end plates and cylinder walls of silver substrates or nickel-buffered copper substrates by a low-pressure plasma-spraying technique combined with a melt-reaction method. The area of the end plate was 177 cm² and that of the cylinder wall was 396 cm². The surface resistance was obtained from the Q values measured before and after substituting either the end plate or the cylinder wall for one covered with the film. The ratio of the surface resistance of the film to that of the copper was lower than 0.08 at 5.2 K and 0.65 at 77.3 K for the end plate and was 0.15 at 5.1 K and 1.91 at 77.3 K for the cylinder wall.

KEYWORDS: high-Tc superconductors, YBaCuO thick films, low-pressure plasma spraying, melt reaction, accelerator cavities, metallic substrates, microwave surface resistance, anomalous skin effect
1 Introduction

The high-$T_c$ superconductors open up many possibilities for practical applications in various fields. The development of synthesis techniques for high-quality materials will accelerate the progress of superconductive microwave applications. A key material property which determines their suitability for practical microwave applications is the surface resistance $R_s$.

For planar microwave and electronic devices such as Josephson junctions and mixers, thin high-$T_c$ films epitaxially grown on single-crystalline dielectric substrates give the lowest surface resistance.\(^1\)\(^-\)\(^4\)

Microwave devices such as antennas, filters, delay lines, couplers and resonators can be made on planar substrates by either thin or thick film processing techniques. However, for some microwave devices such as high-$Q$ cavities for frequency standards, the films must be deposited onto nonplanar substrates with relatively large area. Recently, melt-processed thick films of YBaCuO were fabricated on yttria-stabilized zirconia substrates and were shown to have very low surface resistance.\(^5\)\(^-\)\(^7\) Button and Alford\(^7\) obtained 1.09 m$\Omega$ at 77 K for a 5.66 GHz high-$T_c$ cavity with a total surface area in excess of 230 cm$^2$, which is an order of magnitude lower than the corresponding copper value.

For high-power accelerator cavities, not only must high-$T_c$ films be deposited onto large-area substrates of complex shape, but the use of metallic substrates of high thermal conductivity is also essential. As the thermal conductivity of high-$T_c$ materials is rather low,\(^8\) heat must be released to keep the film in a superconducting state even under a high field. Bohn et al.\(^9\) fabricated bismuth-based thick films (80 $\mu$m) by applying high-viscosity slurries on large-area silver substrates. The sample with surface area of 182 cm$^2$ had surface resistance of 6.7 m$\Omega$ and 9.0 m$\Omega$ at 2.65 GHz and 3.65 GHz, respectively, at 4.2 K.\(^9\) Thallium-based thick films (15$\mu$m) were fabricated by magnetron sputtering onto BaF$_2$-buffered silver-based substrates with surface area of 5 cm$^2$. The obtained surface resistance was 6.9$\pm$2 m$\Omega$ at 11.3 K and 30.2$\pm$1 m$\Omega$ at 77 K at a frequency of 22 GHz.\(^10\) Furthermore, Cooke et al. also showed that deposition onto unoriented silver substrates yielded unoriented films with $R_s$ value of 8.2 m$\Omega$ at 10.6 K and 33.6 m$\Omega$ at 77 K, and that deposition onto oriented silver substrates yielded oriented films with values of 12.6 m$\Omega$ at 11.2 K and 14.6 m$\Omega$ at 77 K at a frequency of 18 GHz.\(^11\) An electrophoretic technique was developed to deposit yttrium-based
thick films (10–20 μm) on silver substrates. A strong magnetic field of 8 T was applied to the suspension to orient the c-axis perpendicular to the substrates. The surface resistance of the sample with the area of 4.5 cm² measured at 21.5 GHz was 18±3 mΩ at 77 K and less than 3 mΩ at 4.2 K.

Plasma spraying can be applied to form high-\(T_c\) films on large-area substrates with complicated shapes. Here we deposited YBaCuO thick films by a low-pressure plasma-spraying technique. In comparison with plasma spraying in open air, it gives a higher density and a better quality of films in a controlled spraying atmosphere. The YBaCuO films prepared by plasma spraying, however, contain some insulating or semiconducting phases such as BaCuO₂, \(Y_2\)BaCuO₅ or CuO, which are precipitated due to the decomposition of \(YBa_2Cu_3O_{7-x}\) during the melting process. This second-phase intrusion cannot be avoided by simply optimizing the spraying or the postannealing conditions. We previously showed that the melt-reaction method reduced the second-phase intrusion and made the \(YBa_2Cu_3O_{7-x}\) layer denser and more uniform.

We measured the microwave surface resistance of the film fabricated by this method. A cylindrical cavity operated at 3 GHz in the TE₀₁₁ mode was employed. The frequency was chosen from the consideration that the fabrication of high-quality films becomes more difficult as the size of the cavity increases, whereas the highest frequency for superconducting accelerator cavities envisaged for the next generation is around 3 GHz. The area of the flat plate was 177 cm² and that of the cylinder was 396 cm². The substrates were either silver or nickel-plated copper. In this report, we describe the sample preparation by the plasma-spraying technique, the method of surface resistance measurement and the results.

### 2 Fabrication of Films

Superconductive films of YBaCuO, the thickness of which is around 30–50 μm, were prepared on silver substrates and copper substrates by a low-pressure plasma-spraying and a melt-reaction method. For copper substrates, nickel was first electroplated to form a buffer layer of about 20–40 μm thickness, which prevents diffusion of copper into YBaCuO films. On the other hand, YBaCuO powder was directly sprayed onto silver substrates. The schematic drawing of the low-pressure plasma-spraying apparatus is shown elsewhere. The powder supplied to the plasma jet was melted and blasted onto the substrates to form the film.
The pressure of the chamber was maintained at around 8000 Pa of oxygen. The feedstock powder was prepared by the solid-state reaction method. Mixed powder of $Y_2O_3$, $BaCO_3$ and $CuO$ with nominal composition of $Y:Ba:Cu = 3:3:4$ (powder I) and $Y:Ba:Cu = 1:13:26$ (powder II) was calcined at 950°C and pulverized. The size of the particles ranged from 26 to 44 μm.

The melt reaction is a combined process of the flux method and the peritectic reaction method. The melt composed of $YO_{1.5}$ and excessive BaO and CuO is in equilibrium with $YBa_2Cu_3O_{7-x}$ and acts as a flux to enhance the grain growth of $YBa_2Cu_3O_{7-x}$. The melt-reaction method consisted of two stages. First, layer I, composed of a mixture of $YBa_2Cu_3O_{7-x}$ and $Y_2BaCuO_5$, is formed using powder I on the substrates by the plasma spraying. Subsequently, layer II, composed of a mixture of $BaCuO_2$, $CuO$ and $YBa_2Cu_3O_{7-x}$, is overlayed using powder II on layer I by the second plasma-spraying process. The thicknesses of layer I and layer II were 150 μm and 50 μm, respectively, in these experiments. In the case of the nickel-buffered copper substrate, the double-layered film is annealed in oxygen at 930°C to 970°C, where layer II melts. The molten oxide diffuses into layer I and acts as a flux which enhances the grain growth of $YBa_2Cu_3O_{7-x}$ in layer I. Simultaneously, the excess flux reacts with $Y_2BaCuO_5$ to form $YBa_2Cu_3O_{7-x}$. These reactions yield a dense and uniform $YBa_2Cu_3O_{7-x}$ layer with thickness of about 40 μm. The excess flux remaining on the reacted layer is removed by polishing with emery paper. In the case of the silver substrate, the double-layered film is annealed in an atmosphere of 7.7% oxygen and 92.3% argon at 900°C for 1 h with a successive heat treatment in 100% oxygen at 700°C for 4 h.

The skin depth $\delta$ is equal to the magnetic field penetration depth $\lambda$ if the temperature is not very close to the critical temperature $T_c$. For single crystals of $YBaCuO$, $\lambda$ at 77 K is about 0.21 μm in the a-b plane and about 1.0 μm in the c direction. The film thickness of over 30 μm therefore ensures that the microwave field does not penetrate into the nonsuperconducting layer to produce additional microwave losses. The microstructure of the oxide layer after annealing was studied with the use of an optical microscope, a scanning electron microscope (SEM) and an energy dispersive X-ray spectroscope (EDX). Figure 1 shows a polarized light micrograph of $YBa_2Cu_3O_{7-x}$. Most of the rectangular grains were about 20 μm long and 5 μm wide. Figure 2(a) is a SEM image of a cross section between the reacted layer and layer I. Figure 2(b) shows the EDX images of the upper middle sextant of Fig. 2(a), showing the distribution of Y, Ba and Cu.
Element analysis revealed that the reacted layer mainly consisted of \( \text{YBa}_2\text{Cu}_3\text{O}_{7-\delta} \) and contained a small amount of \( \text{Y}_2\text{BaCuO}_5 \) and \( \text{CuO} \) grains. The chemical compositions of several points are denoted to the right of Fig. 2(a). The transport critical current density of the superconducting film exceeded \( 2 \times 10^7 \text{ A/m}^2 \) at 77 K. This value is much higher than that of the films prepared by conventional plasma spraying.

3 Experimental Setup

A cylindrical cavity resonating at 3 GHz in the \( \text{TE}_{011} \) mode is used for measuring the surface resistance of the superconductive films. The dimensions of the cavity are 150 mm diameter and 84 mm length. The theoretical values of the copper cavity are as follows. The frequency and the skin depth are 3.02 GHz and 1.20 \( \mu \text{m} \) for both \( \text{TE}_{011} \) and \( \text{TM}_{111} \) modes; the \( Q \) value is \( 4.90 \times 10^4 \) and \( 2.24 \times 10^4 \) for the above modes at room temperature (\( \rho = 1.72 \times 10^{-8} \Omega \text{m} \)). The host cavity was machined out of oxygen-free high-conductivity copper (OFHC) blocks. As shown in Fig. 3, it consists of a top plate, a cylinder wall and a bottom plate. Indium wire of 1.0 mm in diameter was used between the end plates and the cylinder wall for vacuum sealing and electrical contact. The bottom plate has two rf and one vacuum port. Microwaves are loop-coupled into and out of the cavity through two 50 \( \Omega \) coaxial lines. The coupling to the cavity can be varied by moving the line along the axis of the cavity with a fine driving mechanism. The movable length of the line is 25 mm, which enables critical and weak coupling over a wide range of the cavity quality factor (\( Q_0 = 10^3-10^9 \)). The central pumping hole in the bottom plate contributes to breaking the degeneracy between the \( \text{TE}_{011} \) and \( \text{TM}_{111} \) modes. The hole perturbs the field distribution of the \( \text{TM}_{111} \) mode to decrease its frequency, whereas it hardly affects the \( \text{TE}_{011} \) mode. With the pumping hole of 15 mm in diameter the center frequencies of the \( \text{TE}_{011} \) and \( \text{TM}_{111} \) modes become 3.0217 GHz and 3.0182 GHz, respectively, at room temperature. The separation of 3.5 MHz is sufficiently wide compared to the 3dB bandwidth of the \( \text{TE}_{011} \) mode resonance.

The cavity was evacuated with a 10 l/s ion pump and a 50 l/s turbomolecular pump. Typical pressure in the unbaked cavity was \( 10^{-5}-10^{-6} \text{ Pa} \) before cooling. The temperature of the cavity was monitored with thermocouple sensors (Kp vs Au2Fe) mounted on the top and bottom outer surfaces of the cavity. The cavity
The geometrical factor $k$ of the end plate or the cylinder is defined as

$$R_{at}/R_{as}(T) = k(Q_{a,c}(T)/Q_{0,s}(T) - 1) + 1.$$  

$k$ defined below, the surface resistance $R_s(T)$ of the film is given by

$$Q_0:Q_L(l+B_1+B_2).$$

Initially the unloaded quality factor $Q_{0,c}(T)$ of the copper cavity is measured as a function of temperature $T$, and the surface resistance of copper $R_{s,c}(T)$ is calculated. Then, either the top plate or the cylinder wall is replaced by one covered with a superconductive film, and the unloaded quality factor $Q_{0,s}(T)$ is measured. In terms of these unloaded quality factors and the geometrical factors $k$ defined below, the surface resistance $R_s(T)$ of the film is given by

$$R_s(T)/R_{s,c}(T) = k(Q_{0,c}(T)/Q_{0,s}(T) - 1) + 1.$$  

The geometrical factor $k$ of the end plate or the cylinder is defined as

$$k = \int_C H_{\|}^2 dS/\int_S H_{\|}^2 dS,$$
where $H_\parallel$ is the magnetic field parallel to the surface, and the numerator is integrated over the whole surface of the cavity and the denominator is integrated over the surface covered with a superconductive film. For a cylindrical cavity operating in the TE$_{01n}$ mode, $k$ is expressed as
\begin{equation}
k = 2 + \left(\frac{l}{a}\right)^2 \left(\frac{\eta_0}{n \pi}\right)^2
\end{equation}
for an end plate and
\begin{equation}
k = 1 + \left(\frac{2a}{l}\right) \left(n \pi a \frac{\eta_0}{l}\right)^2
\end{equation}
for a cylinder wall. $\eta_0$ is the first root of Bessel's equation $J_0(x) = 0$, and $a$ and $l$ are the radius and length of the cavity.

Now we discuss the error of measurement, where part of the copper cavity is replaced by a superconducting material. As the surface resistance of the material becomes much smaller than that of the copper, the relative error increases because of the difficulty in separating the sample loss from the dominant loss of copper. Differentiation of $R_s$ in eq.(2) with respect to $Q_{0S}$ and additional operations lead to
\begin{equation}(dR_s/R_s)/(dQ_{0s}/Q_{0s}) = -[(R_s/R_s,C) + (k - 1)]/(R_s/R_{s,C}),
\end{equation}
where ($T$) is omitted for brevity and $R_s,C$ and $Q_{0,C}$ are constant. Figure 4 shows the numerical values of $(dR_s/R_s)/(dQ_{0s}/Q_{0s})$ as a function of $R_s/R_s,C$ for $k = 4.09$ (an end plate), 1.96 (a cylinder wall) and 1.32 (the combination of an end plate and a cylinder wall). The error of $Q$ measurement is around 1%. The figure shows that a change of 1% in $(dQ_{0s}/Q_{0s})$ corresponds to about 30% change in $dR_s/R_s$ at $R_s/R_s,C = 0.1$, and about 100% change in $dR_s/R_s$ at $R_s/R_{s,C} = 0.03$ for the end plate. This suggests the necessity of an entire superconducting cavity if we require an accurate value of $R_s$ in a region where $R_s \ll R_s,C$. Another way of improving the accuracy is to measure a mode with a higher frequency. At low temperature the surface resistance of a high-$T_c$ film is approximately proportional to $f^2$ at low field, while $R_{s,C}$ changes with frequency as $f^{0.6-0.7}$. Therefore, $R_s/R_{s,C}$ increases with frequency, making the measurement error smaller.

## 5 Experimental Results and Discussion

In the present experiments, the input power to the cavity is $-14.2$ dBm, where $-4.2$ dBm is caused by the attenuation of the cable connecting the network analyzer and the cavity. From the coupling factor and $Q_0$ of the cavity, the magnetic
field at the cavity surface is estimated to be lower than 0.7 A/m at 4.2 K and lower
than 0.19 A/m at 77 K. It is well known that the rf losses observed in high-\(T_c\) mate-
rials are explained by a model of Josephson coupling between the superconducting
grains.\textsuperscript{26,27} At fields exceeding the lower critical field of the junction \(H_{c1J}\), the rf
magnetic flux penetrates into the Josephson medium and causes rf loss proportion-
tional to the magnetic field intensity.\textsuperscript{28} We confirmed that the surface resistance
is independent of the excitation around this field level and that \(H_{c1J}\) is located at a
higher field.

\section{5.1 Surface resistance of copper}

Figure 5 shows the measured surface resistance \(R_{s,C}\) of the entire copper cav-
ity as a function of temperature. The measured resistivity \(\rho\) of pure copper is
reported in ref. 29. With that value and the permeability \(\mu\), the surface resis-
tance calculated as

\[ R_s = \rho/\delta = (\omega \rho \mu / 2)^{1/2} \tag{7} \]

is plotted in Fig. 5 as well. The measured value at 299.8 K is 15.7 m\(\Omega\), which is
9.9\% higher than that of the theoretical value 14.3 m\(\Omega\). This \(R_{s,C}\) vs \(T\) curve is
fitted by a polynomial of order nine, and is used for the derivation of the surface
resistance of high-\(T_c\) films. In the temperature region below 50 K, the residual
resistance and/or the anomalous skin effect becomes dominant and the figure
suggests that we may assume a constant value for 11.2–4.2 K, a region in which
measurements were not made. We adopt \(R_s = 5.11\) m\(\Omega\), the average of the ten
lowest values.

\section{5.2 Surface resistance of YBaCuO films}

We have prepared seven samples so far: a YBaCuO film on a silver end plate
(labelled YBaCuO/Ag-e), four YBaCuO films on copper end plates (YBaCuO/Cu-
e1 \(\sim\) YBaCuO/Cu-e4 ), and two YBaCuO films on copper cylinder walls (YBaCuO/Cu-
c1 and YBaCuO/Cu-c2).

The surface resistance and unloaded \(Q\) value of YBaCuO/Ag-e are shown in
Fig. 6, as well as those for the entire copper cavity. \(R_s\) is calculated by eq.(2)
using \(Q_{0,s}, Q_{0,C}\) and \(R_{s,C}\). The surface resistance of the sample became equal to
that of copper at 79 K, and less than 0.9 m\(\Omega\) at 50 K. The value near the liquid
helium temperature was 0.1–0.2 m\(\Omega\). For convenience of comparison, we define
the left side of eq.(2) as the normalized surface resistance \( r_n \), i.e.,

\[ r_n = r_n(T) \equiv \frac{R_s(T)}{R_s,C(T)}. \]  

This definition can be extended to include the frequency dependence. The normalized resistance of this sample is shown in Fig. 8 and the following several figures. In Fig. 7, \( dr_n/dT \) is plotted as a function of \( T \). The sharp peak at 91.8 K can be defined as the onset of the transition. Although a high-\( T_c \) film on the silver substrate gave a rather promising result, we chose copper for substrates because copper is more suitable than silver as a basal metal for accelerator cavities in the points of mechanical strength and melting point. Copper is also preferable from the economical point of view.

The normalized surface resistance versus temperature for YBaCuO/Cu-e1 down to the liquid helium temperature is shown in Fig. 8, where \( Q_0 \) changed from \( 1.63 \times 10^5 \) at 5.1 K to \( 1.02 \times 10^4 \) at 118.5 K. The surface resistance of the sample was 1.8 m\( \Omega \) at 5.1 K. The transition temperature was 92 K and the surface resistance of the sample became smaller than that of copper below 80 K. The surface resistance of YBaCuO/Cu-e1 dropped more sharply and reached a smaller value at 77 K than that of YBaCuO/Ag-e. However, the sample exhibited a bending point at 70 K, where the sharp decrease of the normalized surface resistance became approximately constant, indicating that the sample had a rather large residual resistance. The value of \( T_c \) agreed well with the onset temperature obtained from dc resistivity measurement. In the case of YBaCuO/Cu-e2, the behavior of \( Q_0 \) was peculiar and nonreproducible. From the beginning of measurement at the liquid helium temperature, the \( Q_0 \) value was only 12% higher than that of copper. It gradually decreased until 60 K, where it suddenly increased by 20%. It remained near this value until 70 K, then gradually decreased. Since we could not detect any hardware or software problem, we repeated the measurement after twenty-five hours. This time the \( Q_0 \) values were degraded to 67% of the first value at 4.8 K, but again increased slightly at 55 K and followed the same trajectory as the first value above 76 K. As shown in Fig. 9, the transition temperature of the YBaCuO/Cu-e3 sample was almost the same as that of the YBaCuO/Ag-e sample. At 81 K the surface resistance became equal to that of the copper as well. Below 50 K, \( r_n \) became less than one-tenth. The normalized surface resistance from 85 K to 60 K was better than that of YBaCuO/Ag-e. In the case of YBaCuO/Cu-e4, the onset transition temperature was less sharp than in YBaCuO/Ag-e, and above 67 K,
the surface resistance was higher than that of copper. Later the sample was rinsed in acetone bath in an ultrasonic washer to remove the residuals generated in the polishing process. However, this treatment yielded no apparent deviation from the initial measurement. Among the four YBaCuO films on copper end plates, the fourth was annealed in a different furnace from the others, but we endeavored to maintain the same conditions for all samples. As we achieve good reproducibility in fabricating approximately 1-cm-square samples, the dissimilar rf properties of the present samples can be attributed mostly to their size. For larger samples, it is more difficult to maintain the uniformity and stability of temperature and gaseous atmosphere during the heat treatment. A lower temperature or higher oxygen pressure causes an inadequate reaction and leaves residual flux between the grain boundaries.

The surface resistance of YBaCuO/Cu-c1 decreased from 385 mΩ at Tc (91.7 K) to 1.2 mΩ at 5.1 K (Fig. 10). Although the normalized surface resistance was lower than 1.0 below 69.2 K, the absolute value was not as low as those obtained for the end-plate samples. In the case of the second cylinder sample, YBaCuO/Cu-c2, the resonance curve was so broad that we could not separate the TE₀₁₁ mode from the TM₁₁₁ mode. However, the onset of the transition was clearly recognized. To fabricate a better-quality film on the inner surface of the cylinder, the uniformity of spraying and temperature distribution of the cylinder must be improved in the spray process, as well as the uniformity in the heat treatment process.

The best sample, YBaCuO/Cu-c3, showed a sharp decrease of surface resistance below the onset of the transition with a smaller surface resistance than that of copper at 77.3 K. It had a smaller resistance than that of YBaCuO/Ag-e below 85 K, and the surface resistance was less than 0.4 mΩ at 5.2 K, at least one order of magnitude better than that of copper at the same temperature.

6 Conclusions

We have demonstrated that the low-pressure plasma-spraying technique allows fabrication of YBaCuO thick films on copper and silver substrates with large areas comparable to the size of an actual accelerator cavity. The surface resistance was measured at the frequency of 3 GHz using a demountable copper cavity over the temperature range from liquid helium to room temperature. The best film on the
end plate with the area of 177 cm$^2$ had the surface resistance of 4-8\% of that of copper at 5.2 K and about 65\% at 77.3 K. The surface resistance of the film on the cylinder with the area of 396 cm$^2$ was about 15\% of that of copper at 5.1 K. Further improvements in surface resistance and its reproducibility are necessary to fabricate a microwave high-$T_c$ cavity.

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References

Fig. 1. Surface of the YBaCuO film sprayed onto nickel-buffered copper, observed using a polarized light microscope.

(a) Y$_{1.70}$Ba$_{2.34}$Cu$_{3.13}$O$_{x}$
2. Y$_{1.31}$Ba$_{2.53}$Cu$_{3.46}$O$_{x}$
3. Y$_{1.29}$Ba$_{2.56}$Cu$_{3.50}$O$_{x}$
4. Y$_{1.58}$Ba$_{2.37}$Cu$_{3.26}$O$_{x}$
5. Y$_{1.45}$Ba$_{2.44}$Cu$_{3.35}$O$_{x}$
6. Y$_{1.75}$Ba$_{2.26}$Cu$_{3.13}$O$_{x}$
7. Y$_{1.16}$Ba$_{2.60}$Cu$_{3.56}$O$_{x}$

Fig. 2. Cross section of the oxide layer after annealing.
(a) SEM image at the cross section between the reacted layer (upper half of the figure) and layer I (lower half).
The chemical compositions at several points are shown to the right.
(b) EDX images showing the distribution of Y, Ba and Cu in the upper middle sextant of the top picture. The content increases in the order of black, violet, blue, green, yellow and red.
Fig. 3. Demountable cavity for measuring surface resistance.

Fig. 4. Ratio of relative resistance variation to relative \( Q_0 \) variation as a function of the normalized surface resistance.
Fig. 5. Surface resistance of the 3 GHz entire copper cavity as a function of temperature.

Fig. 6. Surface resistance and $Q_0$ value of YBaCuO/Ag-e as well as those of copper.
Fig. 7. Derivative of the normalized surface resistance, showing the onset transition temperature of YBaCuO/Ag-e.

Fig. 8. Normalized surface resistance of YBaCuO/Cu-e1.
Fig. 9. Normalized surface resistance of YBaCuO/Cu-e3.

Fig. 10. Normalized surface resistance of YBaCuO/Cu-c1.