Improved Measurement Accuracy for the Thickness of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ Films by Using a Single Two-resonance-mode Sapphire Resonator

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We measure the thickness of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (YBCO) films at a microwave frequency of 39.91 GHz noninvasively by using the single two-resonance-mode sapphire resonator (TSR) method. In the TSR method, a single sapphire resonator is used for measuring the effective surface resistance ($R_{S,\text{eff}}$) and the penetration depth (λ) of the YBCO films at the same frequency at temperatures of 6 – 88 K. Five YBCO films having thicknesses of 70 – 360 nm are used in this study. The thickness, $t(\text{RF})$, of the YBCO films as measured at 10, 30, 50, and 77 K by using the TSR method turns out to be almost the same as the corresponding one, $t(\text{step})$, as measured by using step profilometry. The differences between $t(\text{RF})$ and $t(\text{step})$ appear to be less than $\sim 5\%$ regardless of the film thickness and the measured temperature, which is smaller than the corresponding value of $\sim 7\%$ as observed by using the two-resonance-mode rutile resonator (TRR) method. The differences between the average $t(\text{RF})$ of each YBCO film and the corresponding $t(\text{step})$ turn out to be less than $\sim 3\%$ in both methods. We discuss the merits in using the TSR method in relation with the TRR method. Our study shows that the TSR method could be very useful for noninvasive measurements of the thicknesses of various superconductor films with accuracy by using microwaves.

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I. INTRODUCTION

The film thickness is one of the most important physical properties of any kind of superconductor films and has been taken into consideration by many researchers in light of the aimed physical properties and fields of applications [1,2]. For homogeneous superconductor films having certain values for the critical current density (J_C) and the intrinsic microwave surface resistance (R_S), the critical current (I_C) and the effective surface resistance ($R_{S,\text{eff}}$) are affected by the film thickness, with the I_C being linearly proportional to the film thickness and $R_{S,\text{eff}}$ depending on the film thickness [3].

Various metrology techniques for measuring the film thickness are known and include stylus profilometry, scanning electron microscopy, transmission electron mi-

croscopy, atomic force microscopy, *etc.* These methods enable us to measure the film thickness directly, which have made them popular for obtaining the thickness of various films despite their invasive nature. There are also some techniques that allow thickness measurements in a noninvasive way, of which four-point probe measurements, X-ray spectroscopy [4], and opto-acoustics [5] are well known for measuring the thicknesses of metal or insulator films. However, these noninvasive techniques have rarely been applied for superconductor films.

Recently, we reported a new method that enabled the thicknesses of superconductor films to be measured noninvasively by using microwaves [6,7]. In the microwave method, the fact of the $R_{S,\text{eff}}$ being described as a function of the R_S and the thickness was used to get the film thickness from the measured $R_{S,\text{eff}}$ of YBCO films having five different thicknesses of 70 – 360 nm at 10, 30, 50, and 77 K by using rutile resonators. The thicknesses of the YBCO films measured at microwave frequencies turned out to be comparable to those from the stylus

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Fig. 1. Step profiles for (a) YBCO-70, (b) YBCO-150, and (c) YBCO-360 as measured by using a step profilometer.

profilometry, with observed differences of $\sim 7\%$ between the two methods regardless of the film thickness and the measurement temperature. One thing to note is that, in measuring the film thickness by using rutile resonators, we had to use a different kind of resonator, *e.g.*, sapphire resonator for obtaining the penetration depth (λ) of the YBCO film samples, because variations in the resonant frequency (f_0) of the rutile resonators with temperature are mostly attributed to those in the permittivity of rutile, making it impossible to get the λ of superconductor films from the temperature-dependent f_0 values. Meanwhile, the permittivity of sapphire is weakly temperature-dependent, which allows us to get the λ of superconductor films from f_0 vs. temperature data for sapphire resonators. We note that information on the λ of superconductor films is essential for extracting the R_S of superconducting films from the corresponding $R_{S,\text{eff}}$ [3].

In this paper, we report measured results for the thicknesses of YBCO superconductor films by using a single sapphire resonator. The two-resonance-mode sapphire resonator method (henceforth called the ‘TSR method’) is used for this purpose [3,8], with the relevant TE_{021} and the TE_{012} resonance modes appearing at 39.91 GHz and 40.02 GHz, respectively. The YBCO films used for this study are the same as those used for the thickness measurements by using rutile resonators. The thickness values measured at the TE_{021} mode frequency are compared with those measured by using step profilometry. The merits of using a single sapphire resonator are discussed.

II. EXPERIMENTAL

Five YBCO films having thicknesses of 70 nm (YBCO-70), 120 nm (YBCO-120), 150 nm (YBCO-150), 280 nm (YBCO-280), and 360 nm (YBCO-360) are prepared on (100) LaAlO_3 substrates by using dc magnetron sputtering. Detailed procedures for the film preparation are

Fig. 2. Diagram of the TSR used for measuring the thicknesses of YBCO films by using microwaves.

described elsewhere [6]. At first, the film thicknesses are measured for identically prepared YBCO films by using a step profilometer. Figures 1(a) – 1(c) show the data collected for YBCO-70, YBCO-150, and YBCO-360 by using a step profilometer.

A cylindrical sapphire rod with dimensions of 5.0 mm in diameter and 2.86 mm in height is used for the thickness measurements by using the TSR method [9]. The diameter and the height of the hollow cylindrical copper cavity surrounding the sapphire rod are 12.0 and 2.96 mm, respectively. Figure 2 shows a diagram of the sapphire resonator used for the thickness measurements. The relevant resonance modes are the TE_{021} and the TE_{012} modes in the two-resonance-mode method, for which the resonant frequencies are 39.91 GHz and 40.02 GHz, respectively.

In using the TSR method, we first get the $R_{S,\text{eff}}$ and the variations in the effective surface reactance with temperature ($\Delta X_{S,\text{eff}}$) from the unloaded quality factor (Q_U) and the temperature-dependent f_0 of the resonator by using the TE_{021} mode, for which the relation between the $R_{S,\text{eff}}$ and the Q_U is described as follows:

$$\frac{1}{Q_U} = \frac{R_{S,\text{eff}}(\text{SC1})}{G_T} + \frac{R_{S,\text{eff}}(\text{SC2})}{G_B} + \frac{R_S(\text{Cu})}{G_S} + k \tan \delta, \quad (1)$$

where $R_{S,\text{eff}}(\text{SC1})$ and $R_{S,\text{eff}}(\text{SC2})$ denote the $R_{S,\text{eff}}$ of the superconductor films placed at the top and the bottom of the resonator, respectively, $R_S(\text{Cu})$ denotes the R_S of copper side wall of the resonator, k is the filling factor, $\tan \delta$ is the loss tangent of the dielectric rod, and G_T , G_B , and G_{SW} are the geometrical factors of the top endplate, the bottom endplate and the side wall, respectively, corresponding to the TE_{021} mode. For reference, G_T ($=G_B$), G_{SW} , and k are 1771 Ω , $2.51 \times 10^5 \Omega$, and 0.9546 for the TE_{021} mode, respectively, with the corresponding values of 585 Ω , $9.78 \times 10^8 \Omega$, and 0.9546 for the TE_{012} mode.

If we use a model equation of $\lambda = \lambda_0 [1 - (T/T_C)^\gamma]^{-1/2}$ for the λ of superconductor films, we get the intrinsic surface impedance $Z_S (=R_S + iX_S = (i\omega\mu_0/\sigma^*)^{1/2})$ from fits of σ_1 and σ_2 in $\sigma^* = \sigma_1 - i\sigma_2$ to the measured $R_{S,\text{eff}}$ and $\Delta X_{S,\text{eff}}$. For YBCO superconductor films, we use $\gamma = 2$. More detailed procedures for obtaining σ_1 and σ_2 of superconductor films have been reported elsewhere [3,9]. Once σ_1 and σ_2 are known for superconductor films, the following relation is used to determine the film thickness

Fig. 3. $\Delta f_0/f_0(T_{min})$ vs. temperature curves for the TE_{021} modes of the TSR and the TRR (inset) having YBCO endplates. Here, $\Delta f_0 = f_0(T) - f_0(T_{min})$, with T and T_{min} denoting the measured temperature and the lowest measured temperature. ‘YBCO’ (open circles) denotes contributions of the YBCO films to Δf_0 , and ‘Rest’ (filled circles) denotes contributions of the remaining parts of each resonator to Δf_0 .

from the $R_{S,eff}$:

$$R_{S,eff} = [\text{Re}(G_h)R_S - \text{Im}(G_h)X_S]. \quad (2)$$

In Eq. (2), $\text{Re}(G_h)$ and $\text{Im}(G_h)$ denote the real part and the imaginary part of the complex correction factor G_h in the relation of $Z_{S,eff} = G_h Z_S$ between the effective surface impedance ($Z_{S,eff}$) and the Z_S . Because G_h depends on the film thickness t , we determine t from a fit of t to the measured $R_{S,eff}$ by using Eq. (1).

III. RESULTS AND DISCUSSION

Figure 3 shows $\Delta f_0/f_0(T_{min})$ vs. temperature curves for the TE_{021} mode of the TSR having YBCO endplates, where Δf_0 denotes the temperature-dependent variations in the resonator f_0 , with $f_0(T_{min})$ denoting the f_0 at the lowest measured temperature T_{min} . In Fig. 3, we see that $\Delta f_0/f_0(T_{min})$ due to the two YBCO films are significantly greater than that due to the rest of the resonator ($\Delta f_{0,rest}$) at temperatures below 50 K, with the two sets of values becoming comparable at temperatures above 60 K. This is how Δf_0 due to the YBCO films ($\Delta f_{0,YBCO}$) can be safely extracted from that of the sapphire resonator, with $\Delta f_{0,YBCO}$ being solely attributed to variations in the λ of the YBCO films with temperature. However, this is not the case for the rutile resonator. In the inset of Fig. 3 for the TE_{021} mode rutile resonator at 15.2 GHz, $\Delta f_{0,YBCO}$ appears to be comparable with $\Delta f_{0,rest}$ only at temperatures below 12 K, with $\Delta f_{0,rest}$ values being $10^2 - 10^3$ times greater than the corresponding $\Delta f_{0,YBCO}$ values at higher temperatures. This is the reason it is impossible to extract $\Delta f_{0,YBCO}$ from Δf_0 . The λ values obtained for the YBCO film samples turn out to be 198, 201, 198,

Fig. 4. Temperature dependences of (a) the $R_{S,eff}$ and (b) the R_S at 39.91 GHz for the YBCO film samples. The inset of (b) shows the calibrated R_S taken at 39.91 GHz as a function of temperature.

196, and 194 nm for YBCO-70, YBCO-120, YBCO-150, YBCO-280, and YBCO-360, respectively.

In Fig. 4(a), we show the $R_{S,eff}$ of the five YBCO film samples measured at 40 GHz. The $R_{S,eff}$ of 1.36 mΩ at 6 K is the lowest for YBCO-360, with the corresponding $R_{S,eff}$ becoming larger as the film thickness gets smaller. The observed dependences of the $R_{S,eff}$ on the thickness are attributed to the interference effects at the film surface between the incoming electromagnetic waves and those reflected at the interface between the film and the substrate [3,10]. The interference effects become smaller for thicker YBCO films due to the exponential attenuation of the electromagnetic fields along the film’s thickness.

The R_S of the YBCO films can be obtained from their measured $R_{S,eff}$ and λ by using the procedures in Section II and are displayed in Fig. 4(b). The R_S values of the YBCO films are comparable throughout the temperatures regardless of the thickness values, which confirms that the observed variations in the $R_{S,eff}$ of the YBCO films, as seen in Fig. 4(a), with thickness are indeed attributed to finite thickness effects. Interestingly, the observed results for the $R_{S,eff}$ and the R_S of the YBCO films imply that the $R_{S,eff}$ of YBCO films can be used to estimate their thicknesses by using the $R_{S,eff}$ vs. R_S relation. In this case, one important thing to consider is that a calibrated data set is needed for the R_S of YBCO films to estimate the film’s thickness from the $R_{S,eff}$ values. Here, we take an average of the R_S of the YBCO films at each temperature and use it as the corresponding calibrated R_S . In the inset of Fig. 4(b), we show the calibrated R_S as a function of temperature.

Figure 5 displays the $R_{S,eff}$ vs. thickness curves, as obtained by using Eq. (1) and the calibrated R_S of the

Fig. 5. R_S^{eff} vs. thickness curves for temperatures of 10 K, 30 K, 50 K, and 77 K at 39.91 GHz, as obtained from the calibrated R_S . The measured R_S^{eff} values of the YBCO film samples are represented by filled circles. The x-axis values corresponding to the five vertical, dotted lines are the pre-determined film thicknesses as measured by using a step profilometer.

Fig. 6. Differences between the film thickness as measured by using a step profilometer ($t(\text{step})$) and that as measured at 39.91 GHz ($t(\text{RF})$) by using the TSR method at temperatures of 10 K, 30 K, 50 K, and 77 K. The dotted lines are guides for the eyes.

$$u_{R_S, \text{eff}} = \left\{ \left(\frac{R_{S, \text{eff}}}{G_E} \right)^2 u_{G_E}^2 + \left(\frac{1}{Q_U} \right)^2 \cdot \left(R_{S, \text{eff}} + \frac{G_E R_S(\text{OFHC})}{G_{SW}} + G_E \cdot k \tan \delta \right)^2 u_{Q_U}^2 + \left(\frac{G_E}{G_{SW}} \right)^2 \cdot u_{R_S(\text{OFHC})}^2 + \left(\frac{G_E R_S(\text{OFHC})}{G_{SW}^2} \right)^2 \cdot u_{G_{SW}}^2 + (G_E \cdot \tan \delta)^2 \cdot u_k^2 + (G_E \cdot k)^2 \cdot u_{\tan \delta}^2 \right\}^{1/2}, \quad (3)$$

where u_{Q_U} , $u_{R_S(\text{OFHC})}$, $u_{\tan \delta}$, u_{G_E} , $u_{G_{SW}}$, and u_k denote the standard uncertainties in Q_U , $R_S(\text{OFHC})$, $\tan \delta$, G_E , G_{SW} , and k , with G_E being a harmonic mean of G_T and G_B and with $R_{S, \text{eff}}(\text{SC1}) = R_{S, \text{eff}}(\text{SC2}) \equiv R_{S, \text{eff}}$ being assumed in Eq. (1). For instance, using the respective relative uncertainties of 2.0% and 2.9% in Q_U [11] and the geometrical factors [12], we get relative standard uncertainties in $R_{S, \text{eff}}$ of 3.5%, 3.5%, 3.6%, and 3.6% at 10, 30, 50, and 77 K, respectively. For this purpose, we use the relative standard uncertainties of 3.1% and 0.01% in $R_S(\text{OFHC})$ and k , respectively, with

YBCO films at 10, 30, 50, and 77 K, respectively. The $R_{S, \text{eff}}$ appears to increase rapidly for thicknesses below 100 nm and to saturate to the R_S as the thickness gets much larger than the corresponding λ values. Using this figure, the thickness of each YBCO film can be estimated by reading the x-axis value, *i.e.*, the thickness value that corresponds to the $R_{S, \text{eff}}$ values measured at each temperature. For instance, the $R_{S, \text{eff}}$ of YBCO-70 is 5.37 m Ω at 10 K, which gives an estimated thickness of 69 nm in the $R_{S, \text{eff}}$ vs. thickness curve for 10 K. We note in Fig. 5 that the estimated thickness values for each YBCO film appear to be comparable regardless of the temperatures. For example, the thicknesses of 69.1, 67.6, 66.8, and 67.2 nm obtained for YBCO-70 at 10, 30, 50, and 77 K, respectively, compared well with the corresponding thickness $t(\text{step})$ of 70 nm as measured by using the step profilometer.

Figure 6 displays differences between $t(\text{step})$ and the thickness $t(\text{RF})$ as measured by using microwaves at 39.91 GHz. The absolute difference between $t(\text{step})$ and $t(\text{RF})$ is less than $\sim 5\%$ for all the films regardless of the temperature, which is smaller than the corresponding value of $\sim 7\%$ measured by using the two-resonance-mode rutile resonator (TRR) method.

The uncertainty in the measured film thickness is directly related with that in the measured $R_{S, \text{eff}}$, for which the standard uncertainty in the $R_{S, \text{eff}}$ ($u_{R_S, \text{eff}}$) of superconductor films can be expressed using the uncertainties in the various parameters in Eq. (1) as follows:

those in $\tan \delta$ being 3.7%, 1.9%, 1.7%, and 7.7% at 10, 30, 50, and 77 K, respectively [13]. The $u_{R_S, \text{eff}}$ values and the corresponding standard uncertainty in the measured film thickness at microwave frequencies ($u_t(\text{RF})$) are displayed in Fig. 5.

Our results show that the two-resonance-mode sapphire resonator (TSR) method has two important advantages over the previously reported TRR method. The first is that the TSR method enables us to measure the thickness of superconductor films by using a single sapphire resonator. This is very important when applying

Fig. 7. R_S^{eff} vs. thickness curves at 77 K as obtained using the calibrated R_S of the YBCO films grown on different substrates. The numbers in parentheses denote the relative permittivities of the substrate materials used for the calculation.

our measurement method for industrial purposes. We note that, unlike the TSR method, the TRR method requires two different kinds of dielectric resonators: The $R_{S,\text{eff}}$ of the superconductor films is measured by using a rutile resonator while their λ values are measured by using a sapphire resonator. Therefore, in using the TRR method, the $R_{S,\text{eff}}$ and the λ are often measured at different frequencies due to difficulties in designing different kinds of resonators with the same resonant frequency. Secondly, the TSR method enables us to realize better measurement accuracy than TRR method does, as mentioned above. This is due to the facts that i) both $R_{S,\text{eff}}$ and λ are measured at the same frequency and ii) only half the number of parameters affecting the measured results in the TRR method are used in the TSR method, thereby enabling reduced uncertainties.

Since our microwave measurement method is based on accurate measurements of the $R_{S,\text{eff}}$ of superconductor films, a physical quantity depending on the reflection of electromagnetic waves at the interface between the film and the substrate, we need to address possible dependences of the $R_{S,\text{eff}}$ of superconductor films on the kinds of substrates used for the film growth. Figure 7 shows the calculated results for the thickness dependence of the $R_{S,\text{eff}}$ of YBCO films grown on five different kinds of substrates, such as LaAlO_3 , MgO , sapphire, Si, and SiO_2 , having different relative permittivity of 24, 9.7, 9.28, 12, and 4, respectively. Interestingly, the dependence of the $R_{S,\text{eff}}$ on the kind of substrate appears to be negligible, verifying the usefulness of the microwave method for measuring the film thickness in a noninvasive way.

Here, we note that our microwave method applies to superconductor films with the same R_S . This may not hold for YBCO films prepared by using different growth conditions and techniques. For instance, the R_S of YBCO films prepared by using the laser ablation method can easily be different from that of YBCO films grown by using the dc-magnetron sputtering method

or the co-evaporation method. Furthermore, even if the same growth method is used for different superconductor films, the R_S of superconductor films depends on the growth conditions, such as the growth temperature, the deposition rate, and the gases used for the growth. In this regard, our microwave method can be best applied to superconductor films prepared by using the same growth technique with the same growth conditions.

We also note that uncertainties in the film thickness as measured by our microwave method are affected by the inhomogeneity along the depth of the film, the surface roughness, the existence of a non-stoichiometric film layer at the interface between the film layer and the substrate, *etc.* In fact, such factors often come into play for very thick or very thin superconductor films, with all of these factors making the R_S greater than the intrinsic R_S value of stoichiometric, homogeneous superconductor films with negligible surface roughness. Therefore, in using our microwave method, it is necessary to check the maximum and the minimum thicknesses of a certain kind of superconductor films under test in advance by comparing the $t(\text{RF})$ with $t(\text{step})$ (or any other thickness values obtainable from direct measurements of the film thickness) for films having various thicknesses. Once the maximum and the minimum thicknesses are known for the kind of superconductor film, the calibrated R_S can be obtained by measuring the R_S of a film having a mean value for the thickness or by taking an average of the R_S of several superconductor films having thicknesses between the two values.

IV. CONCLUSION

We measured the thicknesses of YBCO films at a microwave frequency of 39.91 GHz noninvasively by using the TSR method. A single sapphire resonator was used for measuring the $R_{S,\text{eff}}$ and λ of superconductor films at the same frequency, which made it possible to realize simpler measurement procedures and improved measurement accuracy compared to those in the TRR method. Five YBCO films having thicknesses of 70 – 360 nm were investigated, and the thickness values obtained by using the TSR method turn out to be almost the same as those measured by using step profilometry, with differences being less than 5%. Our study showed that the TSR method could be useful for noninvasive measurements of the thicknesses of various superconductor films by using microwaves.

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