Fabrication of MOD-Derived YBCO Films on (001)LaAlO$_3$ and Their Application to $\lambda/4$ CPW SIR BPFs

Atsushi Sanada, *Member, IEEE*, Masao Kimura, *Student Member, IEEE*, Takashi Yamamoto, *Student Member, IEEE*, and Ikuo Awai, *Member, IEEE*

***Abstract*—Chemically derived epitaxial thin films of $YBa_2Cu_3O_{7-\delta}$ (YBCO) are fabricated on (001)LaAlO$_3$ substrates by the metalorganic-deposition (MOD) process, which has advantages of high quality, nonvacuum, low-cost, and large-scale production of high-T_c superconducting films. The MOD-derived YBCO films have a sharp transition at the critical temperature (90.4 K) and a high-quality film with a surface resistance of 0.13 mΩ (30 K, 9.98 GHz) is obtained. As a microwave application, simple and compact bandpass filters (BPFs) using $\lambda/4$ coplanar-waveguide stepped-impedance resonators are demonstrated on the YBCO films. A two-stage Chebyshev BPF of center frequency of 5.731 GHz, bandwidth of 135 MHz, and insertion loss of 0.29 dB with little input power dependency in a power range less than 10 dBm is realized on the film.**

***Index Terms*—Bandpass filters (BPFs), coplanar waveguides (CPWs), high-temperature superconductors, metalorganic deposition (MOD), stepped-impedance resonators (SIRs).**

I. Introduction

HIGH-T_c superconducting material has immense advantages in microwave applications due to its extremely small conductivity in microwave region [1]. Fabrication of $YBa_2Cu_3O_{7-\delta}$ (YBCO) superconducting thin films by the metalorganic deposition (MOD) process is an approach in which precursor solutions of yttrium, barium, and copper are deposited onto a substrate by spin coating and decomposed/converted to crystalline oxide films of $YBa_2Cu_3O_{7-\delta}$ by heating processes [2]–[8]. Being a nonvacuum chemically derived process, the MOD process have advantages of low-cost and large-scale production of high-T_c superconducting (HTS) films. Also, the MOD approach using metalorganic solution precursors has advantages of precise composition control and rapid deposition rates.

Gupta *et al.* used a precursor system of trifluoroacetate (TFA) salts of the metals [2] to avoid the formation of $BaCO_3$, which is often reported in chemically synthesized YBCO [9] and obtain high-quality films with a critical temperature of 93 K. McIntyre *et al.* investigated growth conditions of $YBa_2Cu_3O_{7-\delta}$ thin films on LaAlO$_3$ substrates and obtained a 70-μm–thick $YBa_2Cu_3O_{7-\delta}$ thin film with a high critical temperature of 92 K with a sharp transition region and a high nonzero field critical current density in excess of 5×10^6 A/m^2 at 77 K [3]. Since then, advancement on the MOD method has been made by extensive works [3]–[8]. Despite of the high potential of the films, their application is mainly focused on power transmission tapes [8] and there is little report on microwave application thus far.

A $\lambda/4$ coplanar waveguide (CPW) stepped-impedance resonator (SIR) is a resonator that consists of two transmission lines with different characteristic impedances and has advantages in size reduction, good spurious characteristics, and low radiation-Q compared with a uniform transmission-line resonator. Having ground planes in the same plane of circuit, the CPW is suitable for a compact $\lambda/4$ resonator with a short circuit at one end of the resonator in terms of easy fabrication, as well as good adaptability for single-side HTS thin films.

In this paper, we fabricate $YBa_2Cu_3O_{7-\delta}$ thin films by the MOD process on (001)LaAlO$_3$ substrates and measured their basic properties including microwave surface impedance at 9.98 GHz. A two-stage Chebyshev bandpass filter (BPF) of $\lambda/4$ CPW SIRs is demonstrated on the YBCO films and a power dependency of the superconducting film is investigated.

II. Fabrication of YBCO Films by the MOD Process

A coating solution was prepared by mixing the acetates of yttrium, barium, and copper in high-purity water with a 1 : 2 : 3 cation ratio with a stoichiometric quantity of TFA at room temperature. The solution was dried by several iterating heat processes adding methyl alcohol to obtain a water- and acetate-free high-purity coating solution [7], [8]. The residue was again dissolved in sufficient methyl alcohol to obtain a spin-coating solution.

The coating solution was deposited on a (001)-oriented LaAlO$_3$ single-crystal substrate by spin coating. The spinning rate and duration was 4000 r/min and 120 s, respectively. The coated sample was fired in a horizontal furnace made of a quartz tube by successive two calcination processes, i.e., decomposition of the solution to an oxyfluoride material at the temperatures up to 400 °C and formation of $YBa_2Cu_3O_{7-\delta}$ at 800 °C.

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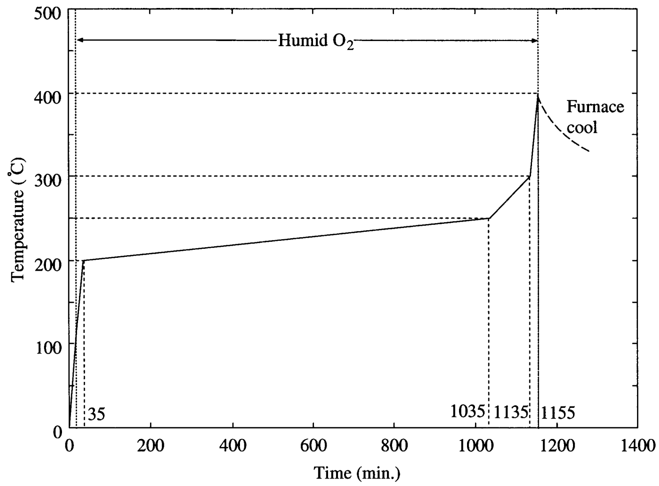


Fig. 1. Heating profile for the calcination.

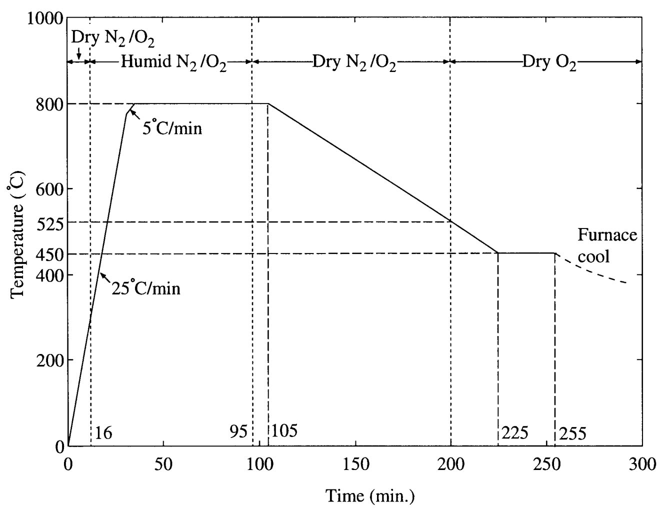


Fig. 2. Heating profile for the annealing.

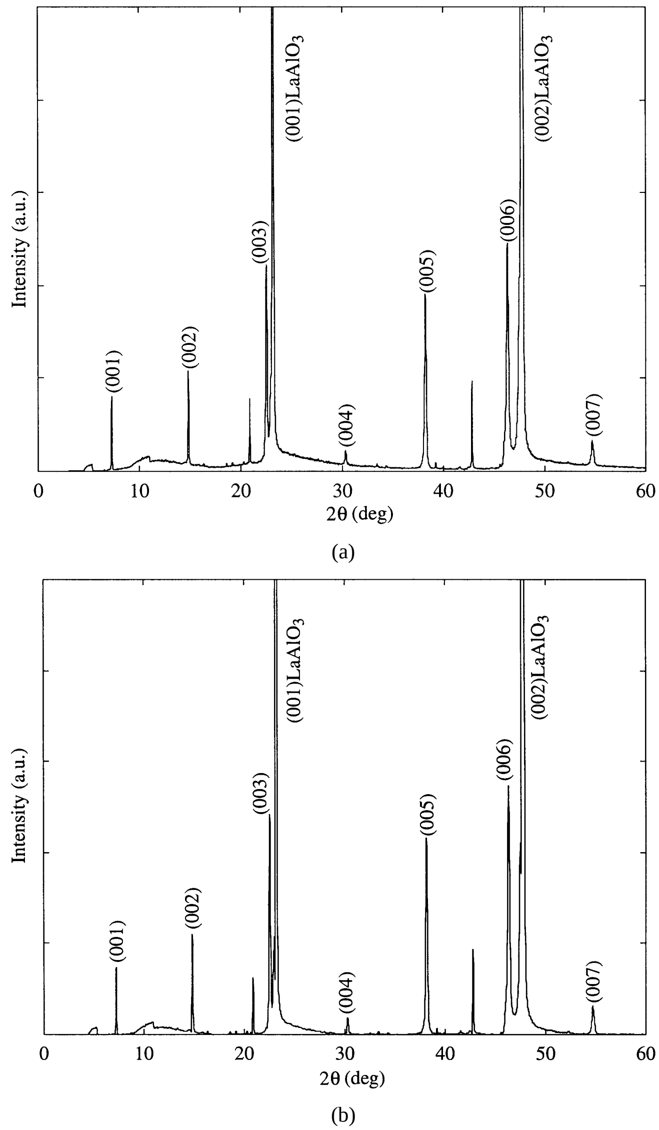


Fig. 3. XRD pattern. (a) $P(O_2) = 0.1$ atm. (b) $P(O_2) = 0.01$ atm.

The temperature profile of the first calcination process is shown in Fig. 1. At temperatures above 100 °C, the furnace atmosphere was kept full of water-contained 1-atm oxygen with a dew point of 25 °C, which is obtained by passing the oxygen through pure water in three flasks in series. The flow rate was set at 1.0–2.0 ℓ/min. The humid oxygen was shut off when the temperature reached 400 °C. The furnace was then allowed to cool in stagnant oxygen. In the second calcination process, oxyfluoride films, which resulted from the decomposition of the metal TFAs, were converted to $YBa_2Cu_3O_{7-\delta}$ in the temperature profile shown in Fig. 2. The furnace was heated at a rate of 25 °C/min to 800 °C. Dry nitrogen/oxygen gas with a oxygen partial pressure of 0.1–0.01 atm was injected into the furnace and switched to humid nitrogen/oxygen gas at the temperature of 400 °C. After a 1—h annealing at 800 °C, the humid gas was again switched to a dry nitrogen/oxygen gas for 10 min and then the furnace was cooled down to 525 °C at a rate of approximately -2.6 °C/min. The furnace was kept 450 °C for 30 min in a dry oxygen atmosphere and cooled to room temperature.

III. Observations of the YBCO Films

A. X-Ray Diffraction (XRD)

In order to investigate an effect of a partial pressure on the orientation of the YBCO superconducting oxide phase, XRD pattern of films annealed in the oxygen partial pressures $P(O_2)$ of 0.1 and 0.01 atm were observed. The results for $P(O_2) = 0.1$ and 0.01 atm are shown in Fig. 3(a) and (b), respectively. The major peaks in the patterns correspond to the $(00l)$ reflections of the superconducting oxide phase, indicating a strong c-axis normal preferred orientation. There is no significant difference between the XRD patterns of the two films in this figure, and it is seen that there is little effect of the partial pressure on the orientation of the YBCO phase for the films of $P(O_2) = 0.1$ and 0.01 atm.

B. Critical Temperature

The resistivities of the films were measured by the direct current four-probe method and the critical temperatures of the

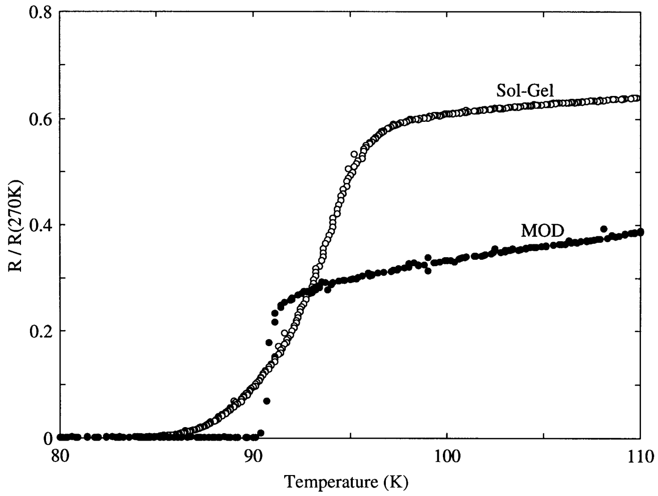


Fig. 4. Normalized resistivity versus temperature. Solid dots are for a YBCO film fabricated by the MOD process with $P(O_2) = 0.01$ atm. Hollow circles are for a typical YBCO film fabricated by the Sol–Gel process derived from the metal naphtenates.

YBCO films were obtained from the zero-resistivity temperature. Fig. 4 shows the normalized resistivity of a YBCO film fabricated by the above-mentioned method with $P(O_2) = 0.1$ atm as a function of temperature. The vertical axis is normalized by the resistivity at 290 K in this figure. As a comparison, the resistivity of a YBCO film fabricated by a typical nonvacuum process of the Sol–Gel process [10] derived from the metal naphthenates are also shown in this figure. It is found that the zero-field critical temperature is 90.4 K with a very sharp resistive transition for the sample obtained by the MOD process.

C. Microwave Surface Resistance

The surface resistances of the YBCO films were measured by the dielectric-resonator method [11] at 9.98 GHz. Fig. 5 shows the measured resistances of YBCO films and their input power dependencies in the power range from -21.7 to 1.67 dBm as a function of temperature. (The input power $P_{\rm in}$ is defined as power flows into the $\rm TE_{011}$-mode dielectric resonator used in the measurements.) Fig. 5(a) shows a resistance of a MOD-derived YBCO film and Fig. 5(b) shows that of a YBCO film made by a typical vacuum sputter process as a comparison. The film fabricated by the MOD method has almost the same resistance as that of the vacuum process, as low as 0.13 mΩ (30 K, $P_{\rm in} = -21.7$ dBm). It is noted that the film also has a sharp transition in the surface resistance and the resistance in the vicinity of the critical temperature (90 K) is better than that of the vacuum process.

IV. $\lambda/4$ CPW SIR BPF Experiments

In order to show a potential of the YBCO films fabricated by the MOD process in the microwave region, a $\lambda/4$ CPW SIR BPF [12] was fabricated on the film and its frequency characteristics of the transmission and reflection were measured.

A YBCO thin film fabricated by the MOD method on a single side of a 20 mm $\times$ 20 mm $\times$500 μm single-crystal $(001)\rm LaAlO_3$ substrate were prepared for the experiment. The relative permittivity of the $\rm LaAlO_3$ is $\epsilon_r = 24.0$. The thickness of the YBCO

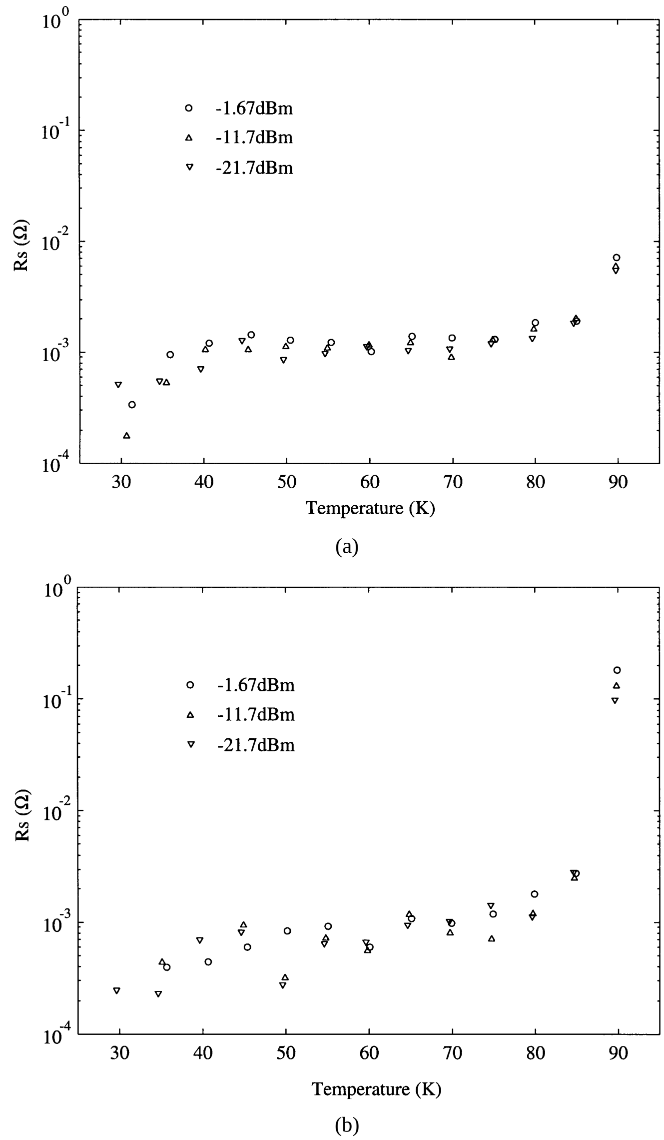
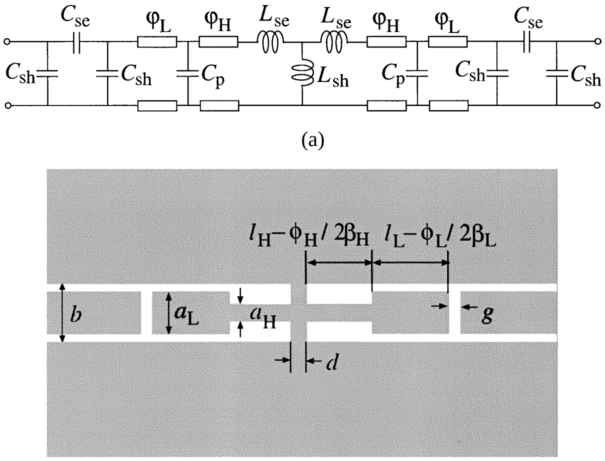


Fig. 5. Surface resistances of YBCO films by: (a) the nonvacuum MOD process and (b) a vacuum sputter process.

film was measured to be less than 0.3 μm. A measured surface resistance of the film is 0.13–0.50 mΩ at 9.98 GHz (input power: $-21.7\sim -1.67$ dBm, 30 K).

A two-stage Chebyshev BPF of the center frequency of 5.8 GHz, the bandwidth of 145 MHz (2.5%), and the passband ripple of 0.01 dB was designed based on an insertion-loss method [13], and precise values of equivalent-circuit elements and the CPW circuit pattern were determined with an assistance of a full-wave electromagnetic (EM) field simulator [12]. The equivalent circuit and circuit pattern are shown in Fig. 6(a) and (b), respectively. Frequency characteristics were calculated by a full-wave EM-field simulator based on the circuit pattern shown in Fig. 6(b). The results of the transmission and reflection characteristics are shown in Fig. 7 with theoretical results obtained by calculating the response of the equivalent circuit of Fig. 6(a), which agree well with each other.

The BPF was set in a package, as shown in Fig. 8, and the transmission and reflection characteristics were measured by using a cryostat below the critical temperature. Input power de-



(b)

Fig. 6. Two-stage $\lambda/4$ CPW SIR. (a) Equivalent circuit. (b) Circuit pattern. $a_L = 0.32$ mm, $a_H = 0.08$ mm, $b = 1.24$ mm, $g = 0.16$ mm, $d = 0.88$ mm, $Z_L = 50\ \Omega$, $Z_H = 73\ \Omega$, $C_p = 0.00882$ pF, $C_{\rm se} = 0.0598$ pF, $C_{\rm sh} = 0.0132$ pF, $L_{\rm se} = 0.0101$ nH, $L_{\rm sh} = 0.0255$ nH, $\varphi_L \equiv l_L - \phi_L/2\beta_L = 1.06$ mm, $\varphi_H \equiv l_H - \phi_H/2\beta_H = 1.86$ mm.

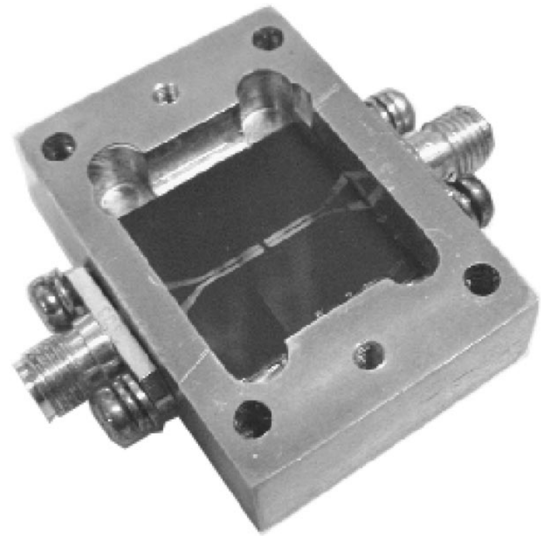


Fig. 8. Two-stage $\lambda/4$ CPW SIR BPF fabricated on a YBCO superconducting thin film on $LaAlO_3$ substrate with a package shield.

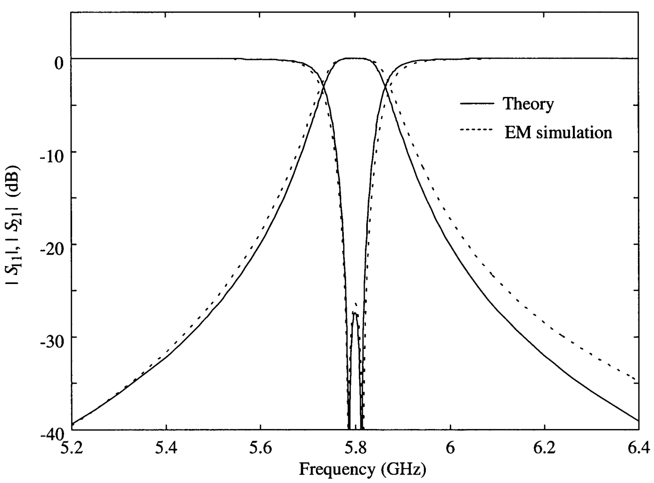


Fig. 7. Frequency characteristics of the two-stage $\lambda/4$ CPW SIR BPF. Solid lines are the theoretical results. Dashed lines are the EM simulation results.

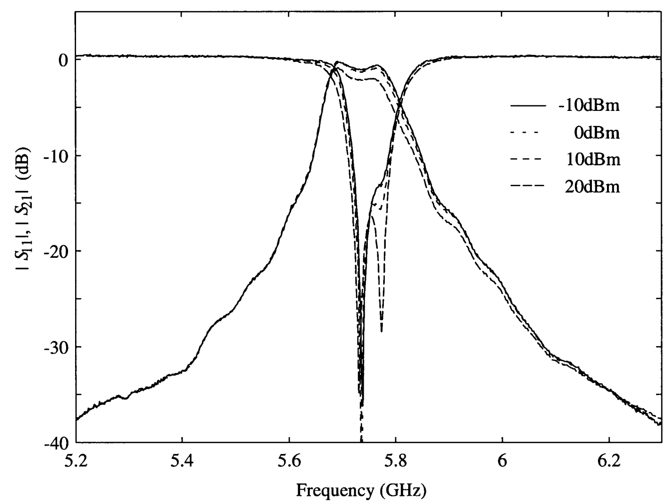


Fig. 9. Measured frequency characteristics of the two-stage $\lambda/4$ CPW SIR BPF fabricated on a YBCO film on $LaAlO_3$.

pendencies of the frequency characteristics were observed in the same time, in the input power range from -10 to 20 dBm. Measured frequency characteristics of $|S_{11}|$ and $|S_{21}|$ at 30 K are shown in Fig. 9. The characteristics agree well with the theoretical results and the BPF with the center frequency of 5.731 GHz, the bandwidth of 135 MHz (2.36%), and insertion loss of 0.29 dB are realized. It is seen from Fig. 9 that the frequency characteristics of the BPF depends little on an input power with a power range less than 10 dBm, but with a power range over 10 dBm, there is conspicuous degradation (typically -1 dB) of the insertion loss in the passband. The maximum permissible power depends on the critical current density and the thickness of the film, which would be improved by optimizing the growth condition of the YBCO film in the MOD process.

Out-of-band characteristics of the BPF are shown in Fig. 10. Spurious transmission characteristics are less than -30 dB within octave frequency range, except a peak at approximately 6.85 GHz, which is considered a resonant peaks due to the incomplete shield package we used.

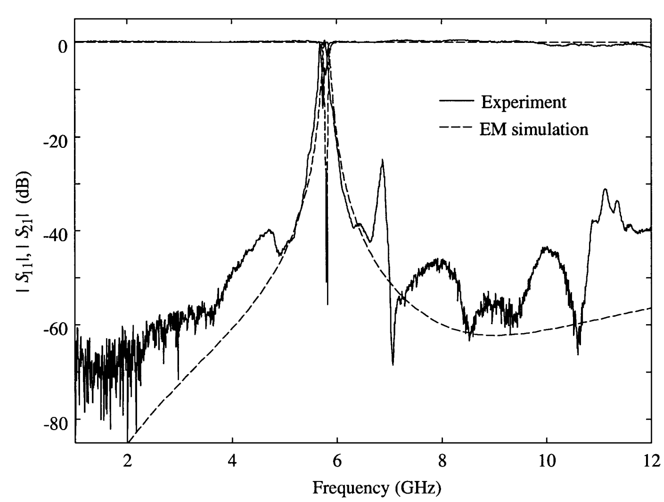


Fig. 10. Out-of-band characteristics of the two-stage $\lambda/4$ CPW SIR BPF.

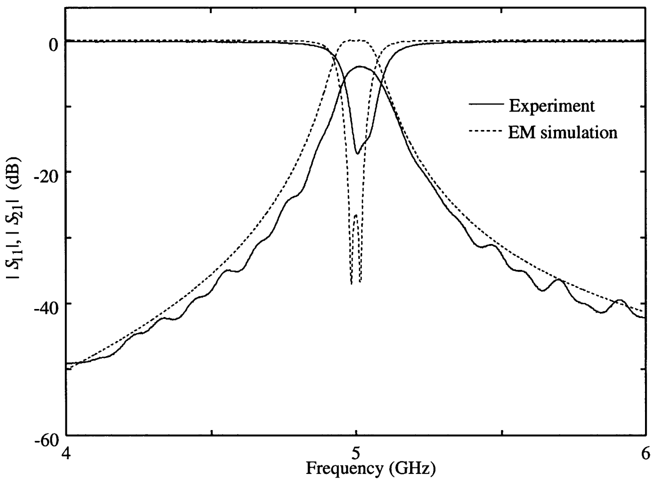


Fig. 11. Measured frequency characteristics of the two-stage $\lambda/4$ CPW SIR BPF fabricated on a normal conductor (Cu).

As a comparison, frequency characteristics of a typical two-stage Chebyshev $\lambda/4$ CPW SIR BPF fabricated on a Cu plated MgO substrate ($\epsilon_r = 9.6$) [12] is shown in Fig. 11. The BPF is designed at a center frequency at 5.0 GHz with a bandwidth of 150 MHz (relative bandwidth of 3.0%) and a ripple of 0.01 dB. Full-wave EM-field simulation results of the BPF based on the corresponding circuit pattern are also shown in this figure. The measured center frequency and bandwidth of the BPF agreed well with the theory and are 5.027 GHz and 155 MHz (3.1%), respectively. However, the insertion loss of the BPF made of Cu is 3.878 dB, which is much larger than that of the YBCO BPF. The advantage of the YBCO films is confirmed.

V. Conclusion

YBCO thin films have been fabricated on $(001)\text{LaAlO}_3$ substrates by using the MOD process, and their microwave surface resistances have measured. The YBCO thin films have shown preferable superconducting property under a critical temperature (90.4 K) with a sharp transition range and a surface impedance of 0.13 mΩ (9.98 GHz, 30 K, $P_{\text{in}} = -21.7$ dB), which is comparable with that made by sophisticated vacuum processes. A compact $\lambda/4$ CPW SIR BPF has been fabricated on the YBCO film, and frequency characteristics and their power dependencies have been measured. A two-stage BPF of center frequency of 5.731 GHz, bandwidth of 135 MHz, and insertion loss of 0.29 dB with little power dependencies in an input power range less than 10 dBm have been demonstrated.

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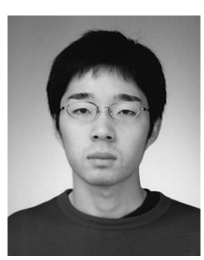

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