Double Sided YBCO Films on 4" Substrates by Thermal Reactive Evaporation

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Abstract—**Homogenous high quality $YBa_2Cu_3O_7$ films on large substrates are first essentials of the commercialization of HTSC devices. We have employed thermal co-evaporation to deposit $YBa_2Cu_3O_7$ films up to 4" diameter. The heater design allows intermittent metal deposition and oxidation in spatially separated zones, with the metal deposition in vacuum and the oxidation at 10^{-2} mbar. On all substrates from MgO, $LaAlO_3$, YSZ, sapphire to Si and GaAs we typically achieved $T_c > 86$ K and $j_c > 2 \cdot 10^6$ A/cm² over the entire area. The radiation heating allows us to turn the substrates over for double sided coating. On 2" and 3" $LaAlO_3$ and on 4" YSZ, both sides meet the above specifications even for thicknesses up to 400 nm.**

I. Introduction

The fabrication of YBCO films of small size up to 2" is now standand in many laboratories, and is near to its commercialization. Larger films of 4" and more, however, are no trivial task as yet, although there is a growing demand. Large areas are simply a must for mass production. They are also required for large electronic circuits like SQUID arrays and transformer coils, for delay lines, filters, and antennas in cellular communications and satellite broadcasting applications, for receiver coils in magnetic resonance imaging, or for multichip modules. Most of these applications require a superconducting ground plane on the back face of the substrate wafer. So double sided deposition is also an important issue on large areas.

A growing demand is further expected in the field of power applications. Particularly promising are fault current limiters of the thin film, resistive type, since they will be compact, fast, and highly economical. Other promising power components may be conductors made on the basis of relatively thick YBCO films deposited on metal tape substrates such as hastelloy or steel, using ion beam assisted deposition techniques. One advantage of such YBCO tapes over BSCCO-"powder in tube" wires is that they are less sensitive to magnetic fields.

In principle, all techniques capable of YBCO thin film deposition can be also used for large areas, by simply scanning the substrate over the deposition region. However, this extends the deposition time and reduces the output of a production facility. For similar reasons, the technique should allow high deposition rates, limited only by the growth process. Other important requirements are homogenous high quality over the full area, and smooth surfaces.

Evaporation techniques are well suited for these purposes, since the deposition is relatively homogenous over large areas. In particular, our technique of thermal co-evaporation[1] allows a close control of the composition via the deposition rates, so that segregation free, smooth films with excellent superconducting properties can be obtained.[2] One problem arose in scaling diameters up, though. The evaporation technique requires ballistic propagation of the metal atoms, and, at the same time, a zone of enhanced oxygen pressure in front of the substrate surface to reactively oxidize the film. This zone has a minimum height which scales with its diameter. For small substrates, the zone is thin, and the metal vapours can easily penetrate. In scaling up, the zone becomes too thick and impedes the ballistic flight of the metals.

We have found a simple trick[3] how to overcome this problem so that we can presently make films on 4" substrates with excellent properties. This will be described in the present paper, together with some recent results, particularly on double sided films, and a few device applications. At first, let us biefly review our technique of thermal reactive co-evaporation.

II. Thermal Reactive Co-Evaporation

Among the various deposition techniques for YBCO, reactive co-evaporation (MBE) is a well established method. While some groups use e-gun evaporators[4] or Knudsen cells[5] as sources, we have chosen boats consisting of current heated metal strips for all three of the metals. These are insensitive to oxygen and allow the use of molecular oxygen rather than atomic oxygen[4] or ozone.[5] This is much simpler, more reliable, and guarantees even oxidation of large areas. The actual arrangement is shown schematically in Fig. 1:

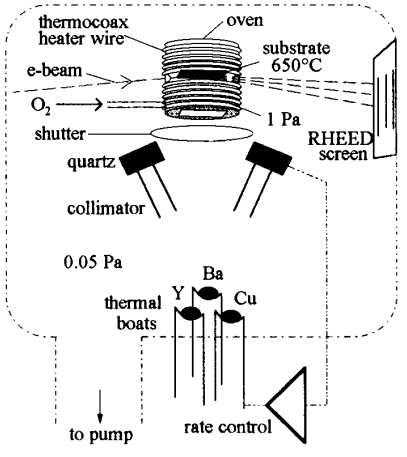

Fig. 1: Thermal co-evaporation scheme

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The heater for small substrates is an oven which heats the sample by radiation from all sides. We mostly use substrate temperatures around 650°C as measured by a thermocouple freely suspended in the oven. We have checked that this temperature agrees well with the actual substrate temperature, as measured by a thermocouple glued into a slit.

The oxygen is fed directly into the oven while the chamber is continuously pumped on, so that a pressure drop from 1 Pa inside to 0.05 Pa outside the oven is established.

Each boat is controlled by a quartz microbalance which has collimators pointing to it. A shutter allows to establish the evaporation rates prior to the deposition of the film. We check the achieved composition mostly just through the surface morphology as observed by SEM, but from time to time also by heavy ion RBS.[2]

On $SrTiO_3$ we have seen many RHEED oscillations, see Fig. 2:

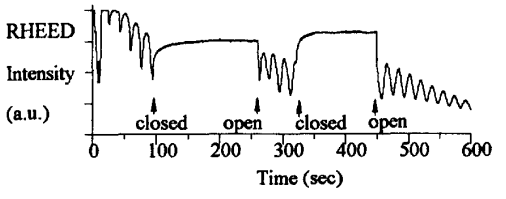

Fig. 2: RHEED oscillations for $SrTiO_3$ substrates. On closing the shutter, the signal recovers with a characteristic time for surface diffusion. On re-opening, the formation of new islands on the terraces leads to a renewed decrease in the oscillation amplitude.

In such experiments we have observed time constants from 0.5 to 20 s, depending on substrate temperature and on the size of the terraces.[6] For the present purpose it is enough to realize that the diffusion times are relatively long, so we can trust that fast oscillations in the oxygen- and metal-supersaturation on the surfaces will be averaged out and will have no influence on the film growth. This is the basis for our trick to sepate the metal deposition and oxidation of the film spatially by a rotating disc scheme.

III. Large Area, Single Sided Films

A. Rotating disc heater

As mentioned above, we had to solve the problem to have a high concentration of molecular oxygen in front of a large surface and yet permit ballistic propagation of the metal atoms to the substrate. Fig. 3 indicates how this was achieved by a rotating sample holder.

Again the substrate is radiation heated from all sides except for the open deposition zone. The oxygen is fed into a separate cup which is open to the rotating wafer but leaves only narrow slits of about 0.5 mm at the sides, so that the oxygen can escape only slowly. By continuous pumping, a pressure drop from 2 Pa inside the cup to 0.01 Pa outside is maintained.

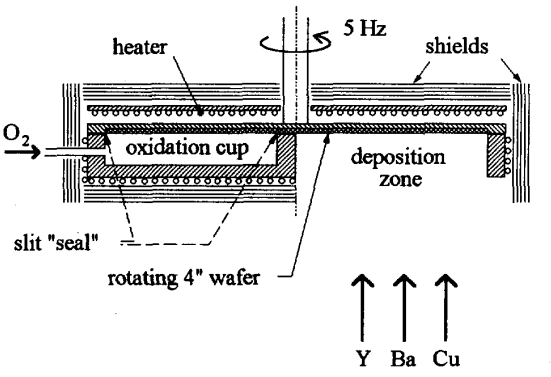

Fig. 3: Heater scheme for intermittent metal deposition and oxidation by a rotating disc sample holder.

The heater is well shielded by a "super insulation" made of thin metal sheets, to avoid excessive heat losses. This way the necessary heater power for 650°C could be reduced to about 800 W, which is basically the radiation loss expected from the Stefan-Boltzmann law for the open area of the deposition zone.

Most substrates require a buffer layer as a diffusion barrier before the superconductor can be deposited. The buffers are usually simple oxides like CeO_2, MgO, and YSZ which can be deposited in molecular form at a low pressure oxygen background by electron gun. For this purpose, we use an extra e-gun in the evaporation chamber which is mounted on a swivel and can be moved in a position just above the boats, underneath the heater. After the buffer is finished, the gun is moved to the side and the boats are used for YBCO deposition without breaking the vacuum.

We often use a fourth boat for an in-situ-overlayer of Au on the YBCO. This way we achieve interface resistances of less than 10^{-8} Ωcm^2.

It is interesting to note the dependence of the film growth from the rotation speed. The effect on T_c is shown in Fig. 4. As expected from the characteristic times observed in Fig. 2, a rotation frequency of 5 Hz is fast enough so that the growth process is not disturbed. At lower rotation speeds the required frequency depends on the deposition rate. About one pass through the oxidation cup is necessary for every 0.1 nm, or each atomic layer.

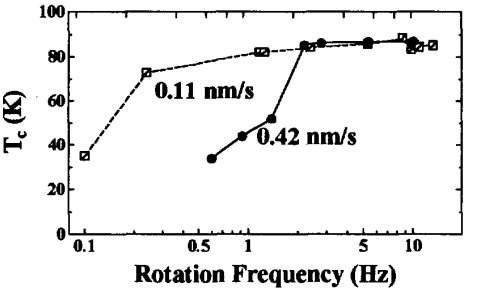

Fig. 4: Dependence of T_c on the rotation speed of the substrate.

B. *Characterization of single sided films*

We measure the superconducting properties, transition temperature T_c and critical current density j_c(77K), of the large films inductively. Excitation and pickup coils are arranged in a transmission setup, and the first harmonic is monitored. We have calibrated the inductive j_c-results by subsequent structuring to microbridges and a resistive measurement. This agrees well with a numerical calculation of the induced currents. In the following Figs 5, we show the results for sapphire and YSZ substrates.

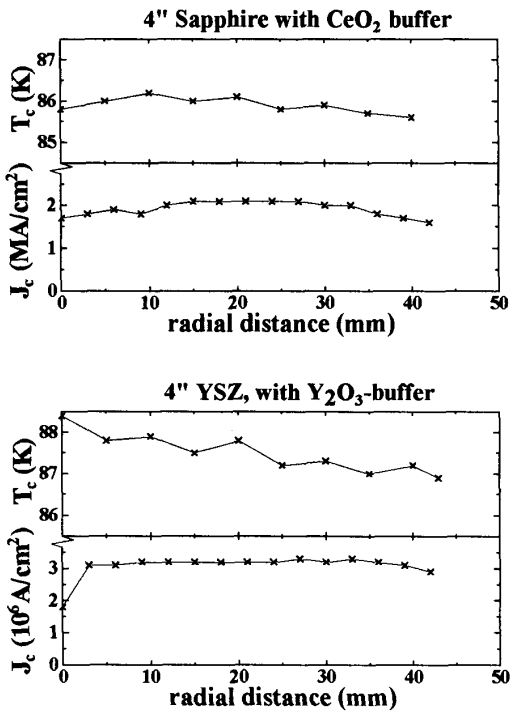

Fig. 5: Superconducting properties of large area films

The inductive measurement is not so well suited near the edges of the films, where it is influenced by feed through, so that the apparent j_c's are sometimes less. But in general the homogeneity is very satisfying. The same holds for the film thickness which varies generally by less than 2%. An exception is a small spot in the center of the wafer with a diameter of 1-2 mm, where the film thickness, and sometimes j_c, deviates because the intermittent evaporation doesn't work there properly.

We can also map j_c(77K) of the full area. We use a simple and inexpensive setup as sketched in Fig. 6. The principle is similar to that of a record player. The liquid nitrogen jar is rotated with the wafer about its axis, and an arm carrying the excitation and pickup coils is swivelled from the center to the rim. The angles are adjusted by computer control on each step so that an orthogonal X-Y-scan is performed.

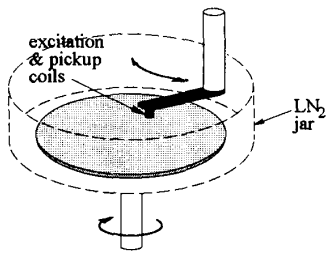

Fig. 6: Arrangement for mapping the critical current density of large wafers.

Since pickup is done on the same side as excitation, we detect the third harmonic here, as a function of the ac excitation amplitude. The apparatus works also for double sided films, because the field is shielded by a factor of 10^4 in the film. A result of a single sided film is shown in Fig. 7:

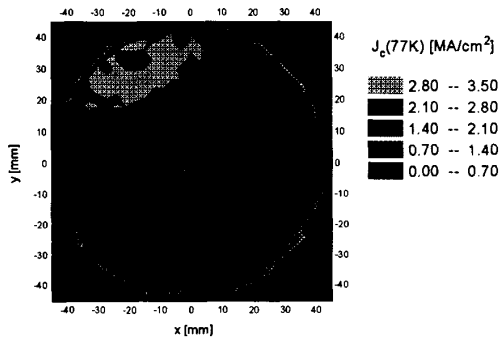

Fig. 7: J_c (77K)-map for a 4" YSZ wafer.

The residual inhomogeneities visible in Fig. 7 are not rotationally symmetric, so that they should be attributed to an imperfect surface finish of the substrate rather than to the deposition process.

A more advanced characterization which is important for many applications concerns the microwave surface resistance. Fig. 8 shows R_s at 94 GHz and 77 K for one of our films on 3" $LaAlO_3$ substrate. The measurement was done by a confocal resonator at Conductus, Inc.[7]

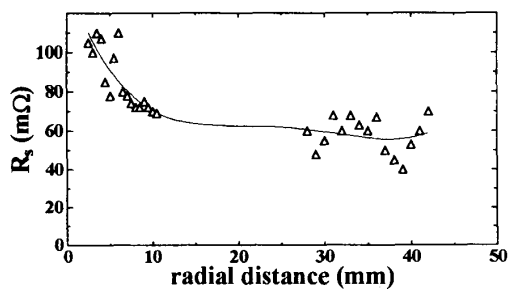

Fig. 8: Surface resistance of YBCO on 3" $LaAlO_3$ at 94 GHz and 77K. Film thickness is 400 nm.

The surface morphology of the large films is practically indistinguishable from that of the small samples, and depends only on the substrate material and the composition. This will be shown further below for double sided substrates.

Apart from the oxide substrates mentioned so far, we have also widely used the semiconductor substrates Si and GaAs, with good success.

C. *Semiconductor substrates*

Semiconductor substrates are partucularly interesting because of the aim to build true monolithic integrated "super-semi" circuits, mainly for RF applications in cellular communications and satellite broadcasting.

Silicon has the additional advantage of being just a cheap high quality substrate available in wafers of 8 inches and more. By using a buffer layer of YSZ to stop diffusion of Si and an additional Y_2O_3 layer to prevent formation of $BaZrO_3$, we were able to make films on Si wafers of 4" with high homogeneity and quality as shown in Fig. 9.[8]

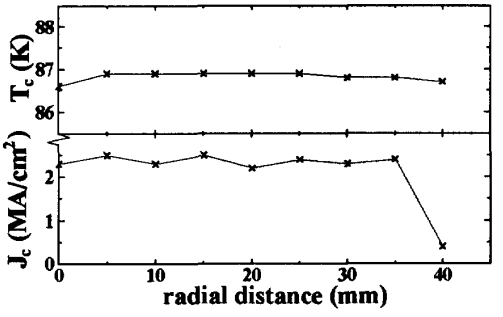

Fig 9: YBCO film on 4" silicon substrate with YSZ/Y_2O_3 buffer.

A problem of Si substrates is the small critical thickness, however. Above a thickness of 70 nm we find cracks induced by the difference in thermal expansion coefficients of YBCO and Si. There are hopes to overcome this problem by deliberately introducing a high density of small pinholes into the film to allow its relaxation, but the tendency is that the superconducting properties become worse.

GaAs has a higher thermal expansion coefficient. However, it seems at first glance totally impossible as a substrate, since it decomposes in vacuum at 490°C already. Fork et al. demonstrated, that MgO can nevertheless be used as a buffer layer, since it grows epitaxially on GaAs at 450°C.[9] However, their j_c was no more than $1.5 \cdot 10^5$ A/cm².

We found out that the reason for this is an arsenic contamination of the YBCO which is not caused by diffusion through the buffer layer but via the gas phase, by decomposition of the wafer on its back face. By passivating all open surfaces of the wafer by a layer of Si_3N_4 we were able to remove this problem and to grow films of standard quality, with $j_c > 2 \cdot 10^6$ A/cm².[10] An example of a 3" GaAs wafer is shown in Fig. 10. We have made films up to 250 nm thickness without cracks.

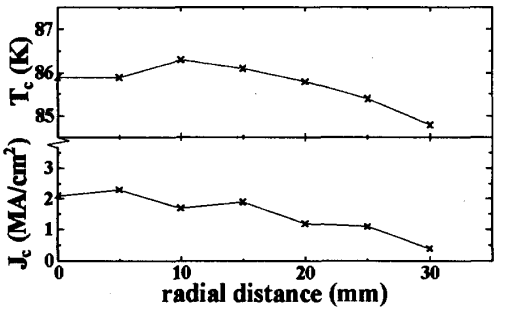

Fig. 10: 3"-GaAs substrate with MgO buffer and a Si_3N_4 passivation layer on all free surfaces.

As already mentioned, many applications require not just one superconducting plane. While we have not worked with multilayer stacks, we have some results for double sided coatings of wafers. This will be addressed next.

IV. Double Sided Films with Large Areas

A prerequisite of double sided deposition is that the wafer must not be glued onto the heater. Otherwise, the first film would deteriorate when the second one is deposited. This is always fulfilled for large wafers, because they would break due to differences in thermal expansion if glued to their support. So the wafer is freely suspended as indicated in Fig. 11, and the heating is done by radiation only.

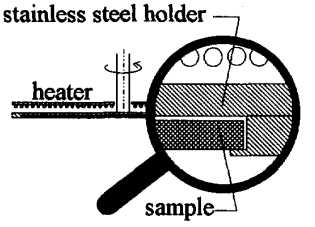

Fig. 11: Suspension of wafer.

So in principle, the wafer can be just turned over and coated again. Some problems arise, though. One is the contamination of the second side by the stainless steel holder during deposition of the first side. We avoided this by simply padding the top side with an extra YSZ wafer, so that the top surface of the sample is exposed to the YSZ rather than to stainless steel.

Another problem comes from polishing. It is difficult to polish the second side of the wafer without at least a bit of degradation of the first side.

A further problem arises when buffer layers must be deposited *in situ* prior to the YBCO films. Then the first YBCO layer may loose oxygen if the buffer of the second layer requires temperatures higer than about 700°C. This problem arose with $LaAlO_3$ substrates which require an *in*

situ-buffer of CeO_2 in order to avoid a-axis grains and a resulting degradation of the critical current density. The oxygen loss can be avoided by feeding oxygen into the heater during the deposition of the second buffer. Also, we found out that the CeO_2 buffers need no growth temperatures higher than 700°C. Fig. 12 shows the j_c- results on 3"-$LaAlO_3$, 400 nm thick.

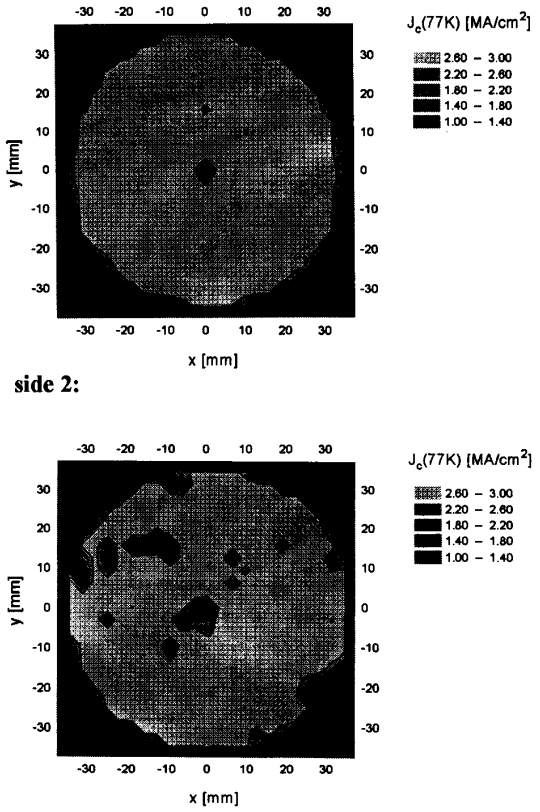

Fig 12: J_c(77K)-maps of double sided coating of a 3"-$LaAlO_3$-wafer with *in-situ*-buffers of CeO_2. YBCO thickness was 400 nm each.

Side 1 is really perfect. It has only the small hole in the center which appears larger here because of the finite spatial resolution of the measurement of a some millimeters. The second side has a few flaws. We believe that these are fortuitous polishing marks. Any contamination would have influenced the wafer as a whole. But clearly, most of the area has very good properties, with $j_c \geq 2.6 \cdot 10^6 A/cm^2$. The corresponding transition temperatures are shown in Fig. 13.

Surprisingly, side 1 has an extremely high T_c which is very homogenous over the entire surface. So it cannot have lost much oxygen during the growth of the second film. Although side 2 has a significantly lower T_c than side 1, it is still rather high compared with our usual results.

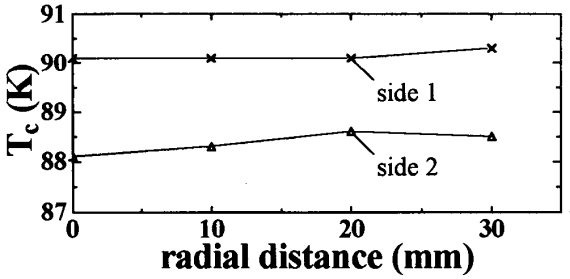

Fig. 13: Transition temperatures for same films as Fig. 12

A further important property of YBCO films is their surface quality. We have used a scanning electron microscope with field emission tip (FE-SEM). The results for another 3"-$LaAlO_3$ wafer are shown in Fig. 14:

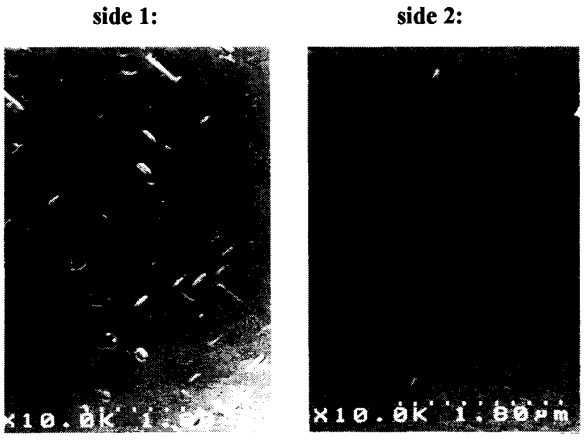

Fig. 14: Surfaces of double sided YBCO on 3"-$LaAlO_3$.

Side 2 is extremely smooth and free of any outgrowth. This is true also at larger magnification and holds for the total area of the film. So the large areas can be made with the same high quality as small areas with our apparatus, indeed. Side 1 has some a-oriented grains. Again the morphology is the same over the whole area. Usually such a-grains indicate a not so perfect surface finish of the wafer. In other cases, we found side 1 to be smoother than side 2. So there is no systematic tendency of the process, and both sides could be made equally well, if the surface finish were equally good on both sides.

As an example of a 4", double sided coating, we show the properties of a YSZ-wafer, see Fig. 15. Again the results are good. Similar data were also obtained for double sided 4" sapphire substrates. So we can say that the difficulties of double sided deposition have been overcome. Before finishing we would like to present a few examples of what the films are used for.

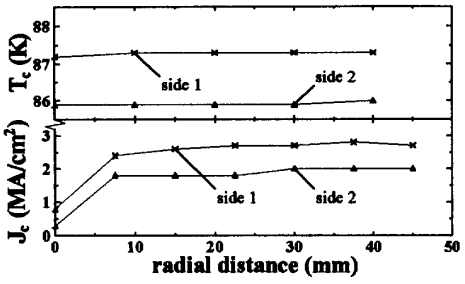

Fig. 15: Double sided 4"-YSZ wafer.

V. Applications of Large Area YBCO Films

While our concern is mostly the fabrication of films, we have collaborations with other groups on various applications. We mention here briefly two examples.

As a first example, Fig. 16 shows a coplanar five-pole filter on a double sided 2"-$LaAlO_3$-wafer made at Conductus[11]

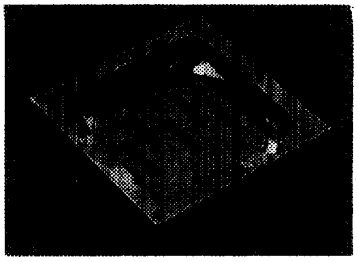

Fig. 16: Coplanar five pole band pass filter for 2 GHz, with YBCO ground plane.[11]

The corresponding filter curve was measured at Lincoln Laboratories.[12] and is shown in Fig. 17:

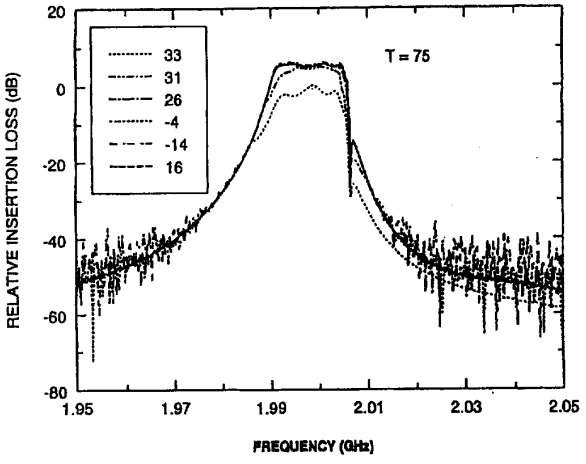

Fig. 17: Filter response with different input powers (dBm), measured with 10 Ω internal impedance at 75 K. Bandwidth is 0.7%.

A second application is a dielectric resonator (sapphire) with superconducting end plates (YBCO on 3"-sapphire substrates) for high power applications made at FIT.[13]. A scetch is shown in Fig. 18, and the extremely narrow resonance curve in Fig. 19.

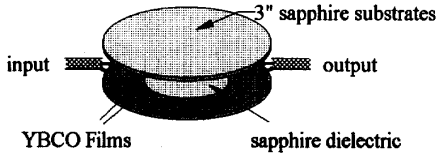

Fig. 18: Dielectric resonator with YBCO end plates.[13]

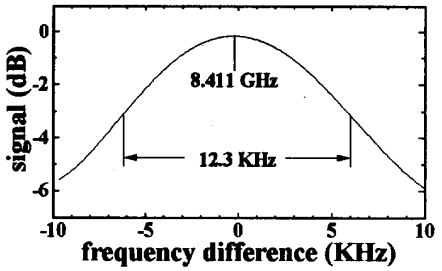

Fig. 19: Response of dielectric resonator with end plates of 3"-YBCO films on sapphire substrates. Intrinsic quality factor is Q_0 = 770000.[13]

VI. Concluding Remarks

We have shown that films with large areas and double sided deposition can be made on the entire area of best superconducting properties and as smooth as the best single films on small substrates. We are presently developing an 8" heater with the same principle.

We are indebted to M. Bauer for help with the manuscript.

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