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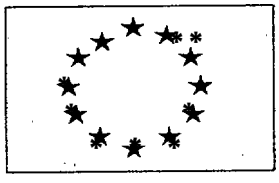

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Development of HTS Thin Films for Microwave Applications

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1 ABSTRACT

This paper reports the achievements of Brite-Euram Project 4283 which had the objective of developing the deposition and processing technologies for t hin film high temperature superconductors (HTS), with an emphasis *on* microwave application ns. Various methods of film deposition and a wide variety of HTS compounds have been invest gated in detail. HTS filters, resonators and delay lines have been characterised and techniq ues for interconnecting and packaging HTS microwave components have been developed.

2 INTRODUCTION

It is widely accepted that one of the first markets for HTS devices w ill be in microwave and high speed electronics applications. Indeed this market is currently expected to be worth 2-3 bn ECU by the year 2000 [1S1S 3]. There is then a good economic justification for developing FITS materials with these applications in mind.

Brite-Euram 4283 has aimed at optimizing the entire process of HTS microwave device fabrication, from HTS powder preparation through to finished devices, including packaging and cooling in a closed cycle cryocooler. Progress for each step in this chain is reported below.

3 TECHNICAL DESCRIPTION

3.1 HTS Powder and Target Preparation

All epitaxial HTS thin film growth methods require a compressed powder target or targets containing either HTS precursor materials or, more usually, HTS material itself. It is normally a requirement that very high target densities are achieved.

Commercial YBCO powder was used in the early stages of the work and the processing conditions for the production of large diameter, dense, high punt y targets were investigated. Solid state reaction synthesis was also used to produce single phase YBCO and ReBCO (where Re rare earth: Nd, Eu, Gd, Er, Dy, Ho) powders starting from $BaCO_3$, CuO and Y_2O_3 or Re_2O_3 Pure, dense targets with good superconductive properties were prepared from these powders. Processing parameters were extensively investigated and were optimi sed.

The main objective of this work was the investigation of the preparation conditions for the production of fully characterised superconducting powders with optimised physiochemical properties. Solid State reaction was used to produce powders of the $Re(Y)Ba_2Cu_3O_{7-x}$ system (Re = rare earth: Eu, Er, Dy, Nd, Gd, Ho).

Re_2O_3 of 99,99'% (Micropure products) or Y_2O_3 99,999% (Alfa products) was mixed with CuO and $BaCO_3$ of 99,999% purity (Alfa products). The mixture was milled in ethyl alcohol with zirconia balls in a Teflon jar. Drying, grinding (with a mortar and pestle) and particle size distribution measurements followed the ball-milling procedure.

Calcination was the next step. This is the most critical stage of the 'whole powder preparation procedure In order to avoid partial melting or considerable grain growth, calcination is carried out at relatively low temperatures (at about 900 "C). At these temperatures the decomposition; of $BaCO_3$ is very slow and is the rate determining step in the formation of the Re(Y)BCO phase.

Thermogravimetric and differential thermal analysis were performed on all the Re(Y)BCO. systems just after the mixing. of the starting oxides. Two endot hermit peaks were mainly observed. The first one appeared at about 820 "C and was due to the structural transition of $BaCO_3$(α to β). The second one appeared at a temperature among 949 "C & 965 "C depending on the powder system and is attributed to the decomposition of $BaCO_3$. A third thermal event at higher temperature (1000 "C) is attributed to the decomposition of the 123 phase to form the 211 one or is due to peritectic melting of 123. phase. $YBa_2Cu_3O_{7-x}$ formation was also investigated by hot stage X-Ray diffraction analysis.

Three different calcination procedures were employed and three different series of powders were produced. The first. one was a four-step calcination procedure (a five-step one in the case of ErBCO system). The powder mixture after grinding was calcined at 900 'C for 10 hours. The whole procedure (ball-milling/ drying/ grinding/ particle size measurement calcining/ characterisation) was repeated four times until the resulting powders consisted of pure ReBCO phase. The fourth calcination step lasted only 7 hours. A fifth step (calcination at 900 *C for 10 hours) was required for the ErBCO system. The other two procedures comprised only one step: calcination at 930 °C for 10 hours and at 950 °C for 35 hours respectively.

X-Ray diffraction analysis of Re(Y)BCO powders showed that the best $YBa_2Cu_3O_{7-x}$, $DyBa_2Cu_3O_{7-x}$ and $HoBa_2Cu_3O_{7-x}$ powders were produced through the four-step procedure comprising calcination at 900 "C for 37 hours. ErBCO system required an extra step (calcination at 900 "C for another 10 hours) for the orthorhombic phase to be completely formed. The intermediate powders produced after each calcination step contained along with the 123 ReBCO phase unreacted Re_2O_3, $BaCO_3$ and CUO as well as the green phase Re_2BaCuO_5. After the first calcination step the 'impurity content was high, but it was consistently reduced with each subsequent step. The final powders were pure, containing the $Y(Re)Ba_2Cu_3O_{7-x}$ orthorhombic phase and only small traces of $BaCuO_2$ or CUO. The only exception was ErBCO powder, which contained some" traces of the green phase. Pure powders of the NdBCO, EuBCO and GdBCO systems were also produced" by the four-step calcination procedure, but the percentage of the tetragonal non-superconducting phase was very high. $GdBa_2Cu_3O_{7-x}$ orthorhombic phase was

completely formed after calcination at 930 'C for 10 hours (me step' procedure), while $EuBa_2Cu_3O_{7-x}$ orthorhombic phase completely formed after calcination at 950 'C for 35 hours. $NdBa_2Cu_3O_{7-x}$ orthorhombic phase was not completely formed u nder any of the processing conditions examined.

Thus it is concluded that each powder system requires different calcination conditions in order to obtain optimum superconducting properties and that the formatio n of the pure orthorhombic phase is especially influenced by the calcination temperature. A high er temperature is required as the ionic radius of the rare earth substituting for Y increases.

The ReBCO powders were used to prepare targets. At first, f or comparison reasons, the research was focused on powders prepared by the multi step procec ure. Various sintering trials were earned out and the properties of the targets produced were measured. For each powder system, an optimum sintering temperature exists where the density re aches a maximum value and the crystallinity as well as the purity of the samples is optimu n. The optimum sintering temperatures were found to be as follows: 990 "C for Dy system, 970 "C for Er and Ho systems, 1000 'C for Nd, Gd, and Eu systems. Sintering at these temperatu es led to maximum density targets. Er and Ho systems exhibit the lowest melting points, thus requiring lower sintering temperatures. Although maximum density was achieved the orthorh mbic phase was not formed in some of these samples. Thus, the conditions for the fabricatio n of targets with optimum superconducting properties were further investigated.

The Y(Re)BCO powders, produced through the optimum calcinat on procedure for each one, were used in this case. Eight different thermal cycles comprising s intering at 930 'C, 935 "C, 950 °C and 980 "C for 10 or 20 hours were investigated in orde to optimise the processing conditions for each powder system. The heating and cooling rate wa s1 °C/min, while annealing was carried out at 450 °C for up to 20 hours. The Tc_{onset} was above 90 K for all ReBCO systems except for NdBC O. These targets are eminentl y suitable for epitaxia 1 HTS Film Deposition

3.2 HTS Film Deposition

3.2.1 Laser Ablation at NKT

At NKT three different methods were applied to upscale laser ablati on film size from 10x1O mm to **2" diameter.** Due to the very forward directed nature of laser ablation there is a significant problem with thickness homogeneity if both the laser beam and the substrate are fixed.

First the laser was scanned on the target surface by computer controlled two axes tilting of the mirror. By this method the deposition profile could be very broad, simply by scanning the laser spot over large areas. However the deposition turned out to be stoichiometric only in the centre of the plume, leading to low quality films in areas grown off-axis. The 'maximum power of the laser decides the maximum target-substrate distance, which at the N KT laser ablation deposition facility was found to be 50 mm. With this short distance between target and substrate the maximum scanning area that could be used, without problems with off-axis depositions, was found to be 12X 12 mm. With a demand on the thickness homogeneity of 10% this limited the

maximum film size to 10x 10 mm. This deposition method was used to deliver 69 10x1O mm films, all fulfilling the initial demands, to the other partners. Preliminary deposition on CeO_2 buffered sapphire has also been performed successfully.

Second it was attempted" to rotate the substrate during deposition. The rotation axis was at the centre of the substrate and by a proper displacement of the plume from the centre of the substrate good thickness homogeneity could be reached on 1 "x 1" substrates. However, the films were not of a good quality because of the off-axis deposition problem. To solve this a mask was placed in front of the substrate to shield large angle depositions. The hole in the mask was made triangular to compensate for the higher tangential velocity of the substrate at the edge relative to the centre. 1 "xl" films could now be made of a satisfactory quality, except at the centre of the film. This was caused by the fact that the alignment of the mask and the film centre was extremely critical because the centre of the film has a fixed position relative to the mask (at the edge of the mask) during the total deposition time.

Finally it was attempted to raster scan the substrate in two directions behind a mask with a 15 mm diameter hole. By this procedure all parts of the substrate experience the same deposition geometry leading to a good homogeneity. The scanning of the substrate was performed with external motors via a bellow. Since also the heating source is placed outside vacuum, this deposition set-up is unique in the sense that no electric feedthroughs are needed. The deposition set-up is shown in fig. 1. 25x25 mm YBCO films have been deposited on $LaAlO_3$ substrates with excellent DC and microwave properties and good thickness homogeneity. In fig.2 is shown the surface resistance versus temperature for a 1 "xl" film. The system is at the moment limited by the area of the substrate holder being 2" diameter and the scanning system which may only be scanned 2" by 2". We have no facility for measuring surface resistance of 2" diameter films, but since 2" diameter films have the same T_c, J_c, thickness uniformity ard surface morphology as 1" x 1" films, it is expected that the 2" films also have state of the art surface resistances. This deposition set-up was used to deliver five 1"x1" and three 2" diarnete r films to ERA.

The surface quality of the films has been improved to the initial stated demand by a reoptimisation of deposition parameters. It was shown that the surface particles are mainly caused by slightly of- stoichiometric films.

3.2.2 **Laser Ablation at the University of Linz**

Investigations were focused on monitoring and improvement of the process and on the comparison of the properties of thin films of different compounds.

PLD was cam-cd out, employing a KrF excimer-laser (λ = 248 nm) and a pulse repetition rate of 10 Hz.' The laser spot on the target was created by imaging instead of focusing in order to an inhomogeneous distribution of energy density in the laser spot. Imaging of circular apertures permits to vary the spot size fast and accurately. The deposition parameters were optimised, employing home-polished (100) MgO substrates and Y-123 targets We found the following set of parameters to be suitable for producing Y-123 films showing good superconducting properties: Substrate temperature T = 750°C, oxygen partial pressure p(O2) = **0.7** mbar, laser

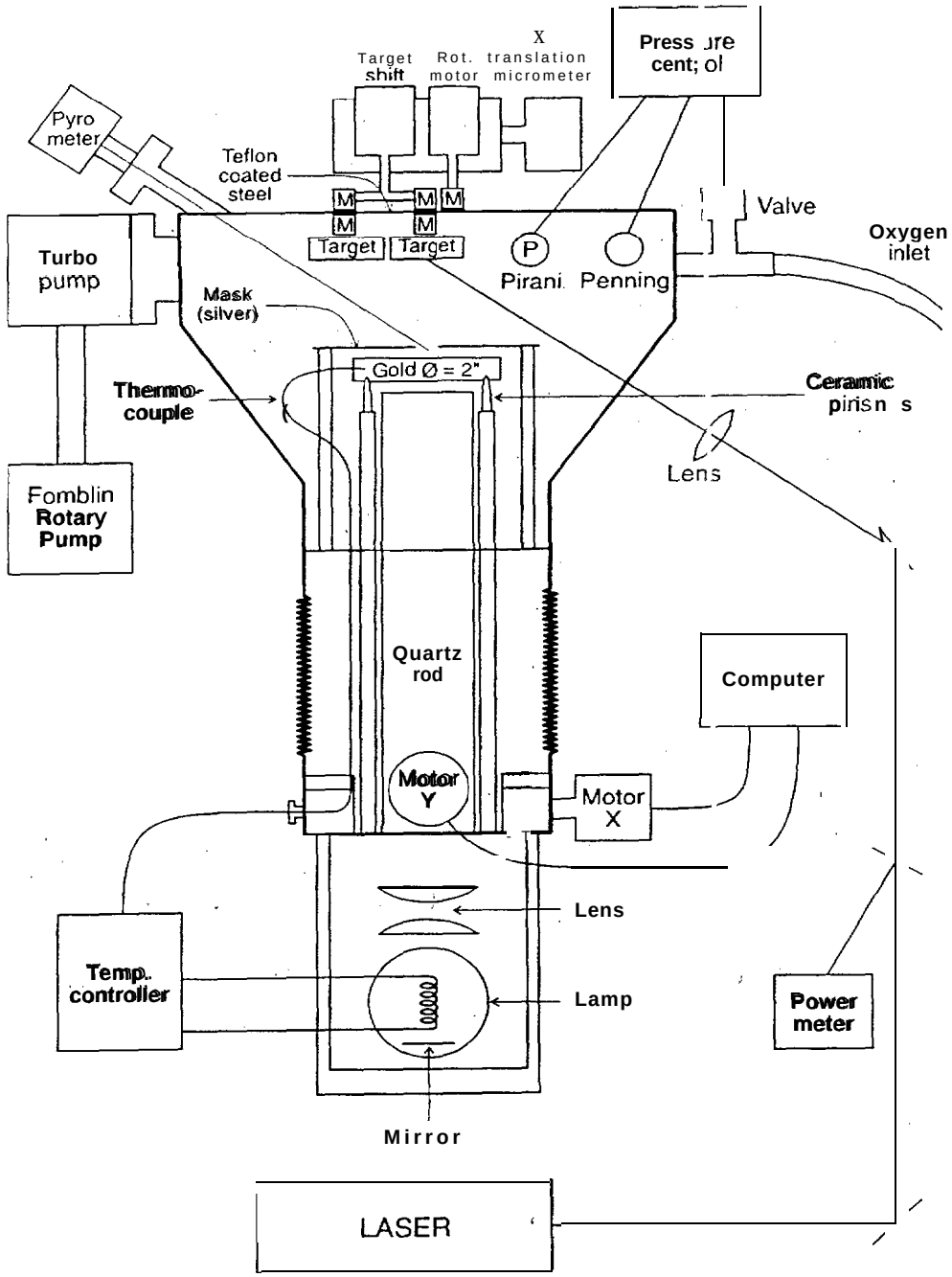

Figure 1: NKT Film deposition equipment

energy E = 80 mJ, spot size A = 2 mm2 (F ⁼ 4 J/cm2), and targ >t-substratedist.ante d = 5 5 mm.

To monitor the growth of BISCCO thin films a facility has been set up to **measure** the reflectivity of the substrate throughout the whole process. The He-Ne-laser beam ‹s partly reflected on the front side and partly transmitted and reflected on the back side. The optical path difference DL" of the interfering beams depends on the refractive ind :x of the substrate, n, on its thickness, d, and on the angle of incidence, q. Both n and d depend on temperature. When the substrate expands while increasing temperature, the changes in DL cause a periodic change in

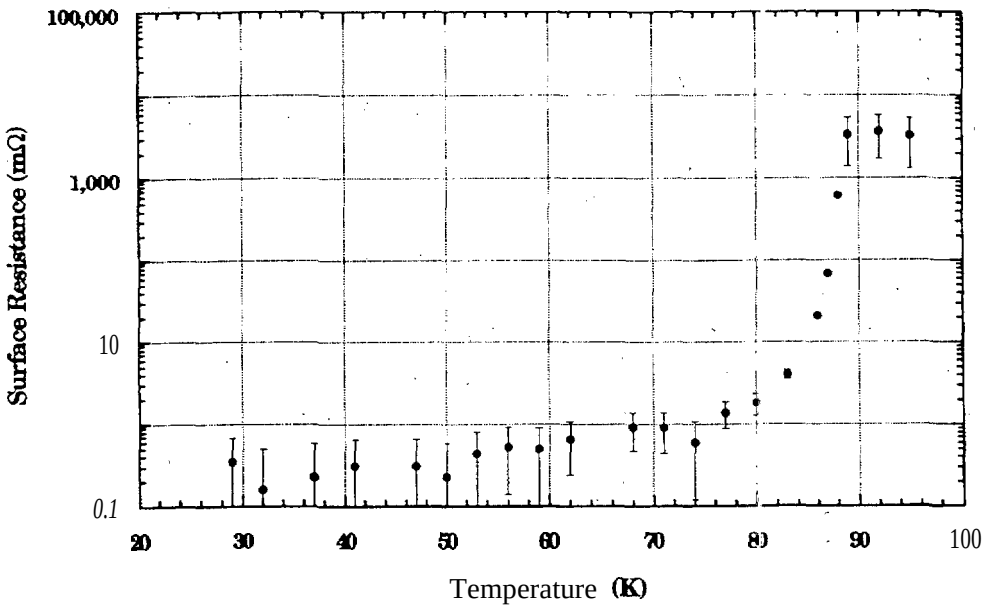

Figure 2: Surface resistance versus temperature for a 1 "xl" YBCO film

the measured intensity of the reflected light. It is' possible to mess ure the temperature with an estimated accuracy of ±3°C at T = 800°C.

3.2.3 MBE **at the Technical University of Denmark**

During the project we have produced high-quality superconducting{; $YBa_2Cu_3O_{7-x}$ thin films on single sided and double sided 10x 10 mm^2 and 25x25 mm2 $LaAlO_3$ substrates. Also, we have demonstrated that the process can be extended to include 2 inch subs tates.

By applying a double deposition method (with double annealing) with each YBCO layer being 150 nm thick, the film surface morphology was more smooth and the critical current densities were improved.

The work has exclusively been concentrated on the BaF_2 process. First, using an MBE technique Y, BaF_2, and Cu are co-deposited at room temperature on the substrate that will hold the superconductor. Secondly, postannealing with oxygen and water in a furnace substitutes the fluorine with oxygen, thus producing the superconducting $YBa_2Cu_3O_{7-x}$.

Controlling film stoichiometry is an iterative process in which a trial film is produced and subsequently chemically analysed. From the chemical analysis the next deposition can be adjusted accordingly to obtain good stoichiometric composition. It is necessary to use an iterative process for finding the correct evaporation rates for each element because no good technique for monitoring the absolute amounts of each clement is available. Stoichiometry 'fine tuning' is also necessary each time the MBE system has been reloaded with fresh source metals (Y, Ba, Cu).

After reloading source materials, the MBE system can produce approximately 10 batches of superconducting YBCO thin films.

After the ex situ deposition process was optimised, the annealing conditions were optimised for maximum quality of the thin films. An optimum annealing profile was found after elaborate testing of temperature profile, step time and pressure. At first an oxygen pressure of 1 atmosphere was used at the maximum profile temperature but from work of Mogro-Campero (A. Mogro-Campero, L. G. 'Turner, *Appl. Phys. Lett. 58 1 417)* it was realised that the oxygen pressure had to be lowered in order to synthesise a proper YBCO phase. After extensive optimisations, an optimum was found at 800°C and 1.0 mbar partial pressure of oxygen.

A major problem in growing thin films with the BaF_2 process and ex-situ annealing is that the critical current density decreases as the film thickness is increased. Thus, if the current transport capacity of the film is of major concern then the growth process (i.e. annealing) should be further optimised in order to overcome this problem. Also, the surface morphology gets worse with increasing film thickness. This has a negative impact on *the* post-growth processing and, thus, the reliability of the devices. Data from a visit at the Physics Department, University of Rome (Italy) clearly showed that the top layer of the YBCO film is a-axis oriented when the film thickness exceeds a certain limit (200 nm) using the BaF_2 process with ex-situ annealing).

In order to overcome both the surface morphology problem and the problem with the electrical properties, both caused by the a-axis growth probability for thicker films, a double layer deposition process was tested. In the first experiment" a 2 x 200 nm film was grown by first growing a 200 nrn film, then anneal using the optimum annealing profile (8 OO"CI1OO Pa/l hour+525°C/1 atm/1 hour), then growing a second 200 nm film on top of the first and once more perform the, annealing. Thus, the bottom 200 nm film was annealed twice. The results were somewhat disappointing in that Jc was low (5x105 A/cm^2) and the surface morphology was poor. We then concentrated on the annealing sequence and furthermore reduced the thickness of each layer to 150 nm. For the annealing sequence we tested the effect of.> reduced annealing time for each layer. We found that reducing the annealing time for each layer in the high temperature step (800°C/100 Pa) to 15 minutes, while holding all other parameters constant, gave properties almost as good as for the 150 run films annealed using the optimised annealing sequence. The surface morphology was good and the value of J_c was only slightly [Deteriorated (from 4.5x1 0^6 to 3.5x1 0^6 A/cm^2). However, reducing the, annealing time in the low temperature step (525°C/1 atm) to 15 minutes, while holding *all* other parameters constant, was found to have a more negative impact on J_c (from 4x 10^6 A/cm^2 to 1.4x10^6 A/cm*). The surface morphology, again, was good.

In order to fabricate substrates with $YBa_2Cu_3O_{7-x}$ thin films "on both sides we use a special substrate holder with a hole in the centre. After deposition of $Y(BaF_2)_2Cu_3$ on one side of the substrate, the sample is turned upside down in the MBE system load-lock and recentered into the MBE system main chamber for deposition on the other side. The whole process is quick and easy using the special substrate holder. When $Y(BaF_2)_2Cu_3$ has been "evaporated on both sides, the sample is *ex situ* annealed in a furnace.

The BaF_2 process has some technical advantages to the *in situ* process: There is no oxygen present in the chamber during deposition. Oxygen affects the surface of the evaporation sources and makes the metal fluxes dependent on the oxygen pressure. Using BaF_2, the sample is heated in an oven at a one atmosphere pressure where thermal contact is accomplished through convection. *In situ* temperature control over a large area is not as easy as it might sound. At low pressures radiation and thermal conduction are the only ways to transfer heat to the substrate. Inhomogeneous heating is often the result.

3.2.4 **Sputter Deposition at Demokritos**

The deposition of $YBa_2Cu_3O_{7-x}$ (YBCO) in situ, by a sputtering technique has proved to be one of the most best techniques for depositing high quality HTS films. The term in situ means that YBCO phase is formed during growth. The formation takes place or the heated substrate surface from an incoming flux of atoms, except for the final oxygen occupancy in the structure which takes place during the cool-down (annealing). In contrast, in the ex -situ method, an amorphous or polycrystalline film is deposited first, and a post deposition process is required after, which includes heating near the melting point in order to have crystallisation of the film, starting at the interface with the substrate. As a rule, thin films grown in situ are superior to ex situ films in terms of epitaxy, surface smoothness, reduction of substrate interdiffusion and high J_c values. The main problem of the deposition of YBCO in situ by a sputtering technique, is resputtering Resputtering occurs when high energy, negatively charged particles (mainly 0-) **which are formed on the cathode, accelerate towards the** anode (heater surface), impact upon the growing film and cause compositional changes (mainly Cu and Ba are preferentially etched). The result is a multi-phase film with poor superconducting properties. In order to overcome this problem, three main techniques are used.

The first is to use a non stoichiometric target in order to compensate for the stoichiometric changes because of the resputtering effect. This technique requires the use of RF sputtering because the non-stoichiometric targets are insulators, and considerable time and effort to find the proper target composition.

The second is to sputter (mainly with DC excitation) at high total pressures ($Ar+O_2$ over 400 mTorr) from a stoichiometric target, in order to reduce the kinetic er ergy of the particles because of the very short mean free path, and as result reduce resputtering. This approach also enables the use of high partial pressure of 0_2 during growth.

The third technique is to change configuration and to place the substrate on the side of, and at an angle to, the sputter gun (more or less 90°). This is termed off-ax is sputtering. In this way the growing film is out of the stream of the energetic particles which strike on the substrate. However this method results in low deposition rates and poor thickness uniformity.

The last two techniques seems to be the most favoured ones for the deposition of high quality YBCO thin films by a sputtering technique. It is important to note that sputtering process has many technical peculiarities (magnetron sources, surrounding configuration of the chamber etc.) that influence the deposition process and differ from system to system. We chose to sputter in an

on-axis in our system because the thickness uniformity was poor i ı the off-axis configuration. The parameters that had to be optimised in order to find the proper conditions of deposition are the following: 1) The total pressure of the mixture of gases ($Ar+O_2$) in conjunction with the gas flow ratio (flow O_2 / flow Ar) in order to minimise the resputtering effect and maintain enough partial pressure of 0_2 during growth from the other, which permit the formation of YBCO phase. 2) The proper substrate temperature, which results in the epitaxial growth of YBCO thin films with superior properties and depends on the substrate used. 3) The power supply of the target in conjunction with target to substrate distance, which determines (depending on the pressure) the deposition rate and the energy of the species that reach the substrate. We used both RF and DC excitation of the sputtering source.. We used two new methods in the sputtering of YBCO. One was the use of a *powder target* and the other was the *insertion of conducting disk* between **target and substrate.**

The resputtering problem can be solved with the use of a conducting disk between the target and the substrate. This disk plays the role of the collector of the negatively charged ions and only permits particles with sloping trajectories to pass to the substrate. These particles have lost a large part of their energy due to the larger distance which they have to cover. Consequently, the resputtering phenomena was reduced drastically y. In this configurate ion we placed a copper foil with disk shape between substrate and target. In this stage of the work we returned to the sintered targets after solving the cooling problems in the mounting of the target. The results had better reproducibility and the superconducting properties were very good. We obtained the best result at 15 mTorr O_2. The use of the disk gives films with better surface morphology in respect to those which have been deposited without the disk. With this method we are able to produce thin films with acceptable superconducting and surface morphology properties but with a limitation in the size of the films.

In order to successfully deposit larger area we changed the conditior .s of the deposition. The new conditions are DC-magnetron sputtering in ON-axis configuration.

Using stoichiometric targets with resistivity <40 (Ohm/cm) we succeeded in depositing thin film $YB_2Cu_3O_{7-x}$ with good superconducting properties up to 1" x 1". This method is promising because its reproducibility was better than the previous RF-sputtering. We have succeeded to deposit thin films with T_c up to 90K. For example, two films 1" x 1" have T_c=86K and J_c=4.5MA/cm^2. Deposition of large area $YBa_2Cu_3O_{7-x}$ thin films leads to films with a large T_c but also to non uniformity so that the films have ATC=5K.

3 . 3 **HTS Microwave** Device Fabrication **and Testing**

Films produced within the project *were* processed into devices at ERA and then tested and characterised to provide feedback for film quality improvement Coplanar and microstrip resonators and "delay lines and microstrip filters were produced.

The initial resonator design used in this work was a microstrip ring resonator (to reduce radiation losses) with a gold ground plane, capacitively coupled via a sma l gap to 50 S2 transmission

lines. As film quality improved however, the gold resistance became a limiting factor on device performance and the design was replaced with a coplanar linear structure.

The original design for the delay line employed a 50 microstrip folded on itself to give maximum length on a 10 mm square substrate. A device made with a gold microstrip exhibited a trough of high loss centred at approximately 4.8 GHz. This effect was attributed to an interaction between adjacent sections of the microstrip, with the resonant frequency determined by the length of the straight sections.

Meandering each bank of the delay line five times gave good performance up to 10 GHz and use of tapered impedance transformers enabled longer delays to be fitted onto the tile. However in order to produce long delays large double sided wafers would be needed. At this time, development of large area (2 inch) films by NKT was progressing. The shape of the 2 inch substrates was circular as opposed to the square shape of the smaller substrates. In order to use this shape efficiently, the delay line design was at the same time changed to that of a double spiral. Figure 3 shows the design for a 1 inch HTS film. Designs for 2 inch and 10 mm films were also produced.

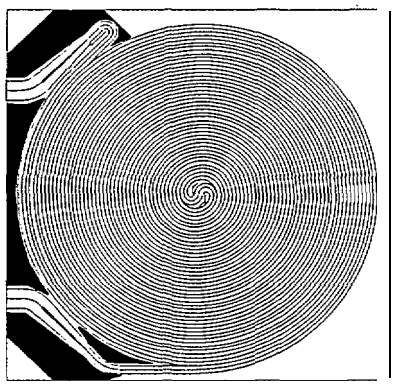

Figure 3: Mask, pattern for 25mm delay-line

One three pole Chebychev filter was designed in order to allow comparison of a 'real' HTS microwave component with its gold counterpart. It also enabled a comparison of design predictions with an actual, device.

Fabrication of devices. began with removal of silver epoxy used for heat transfer during film growth. Before patterning of the HTS *film* it was selectively coated in 1μm gold layers by laser ablating gold through a shadow mask. Contact pads were formed from these gold areas by patterning them using KI:I gold etch, after the HTS film had beer patterned. The completed device was then attached to a carrier. Early devices were fixed *to* brass carriers by sliding clips to allow for differential expansion on cool-down. Later in the project, the brass was replaced

with kovar alloy, which has a better expansion match to lanthanum aluminate (and alumina) and the device was attached with non-corrosive silicone elastomer. The later design was easier to assemble and more robust. Connection to the HTS device was made with a sliding pressure contact for the early devices and later with conventional wedge-bonding.

3.4 Cryocooled Device Fabrication

Following the 6th progress meeting it was agreed that Oxford Instruments would source a Stirling cryocooler for incorporation into a self contained 19 inch rack unit as a final demonstrator unit.

An appropriate 500 mW at 80K Stirling cryocooler was found and placed on loan with Oxford Instruments by Hymatic Ltd for six months. Calculations were done to estimate the heat load

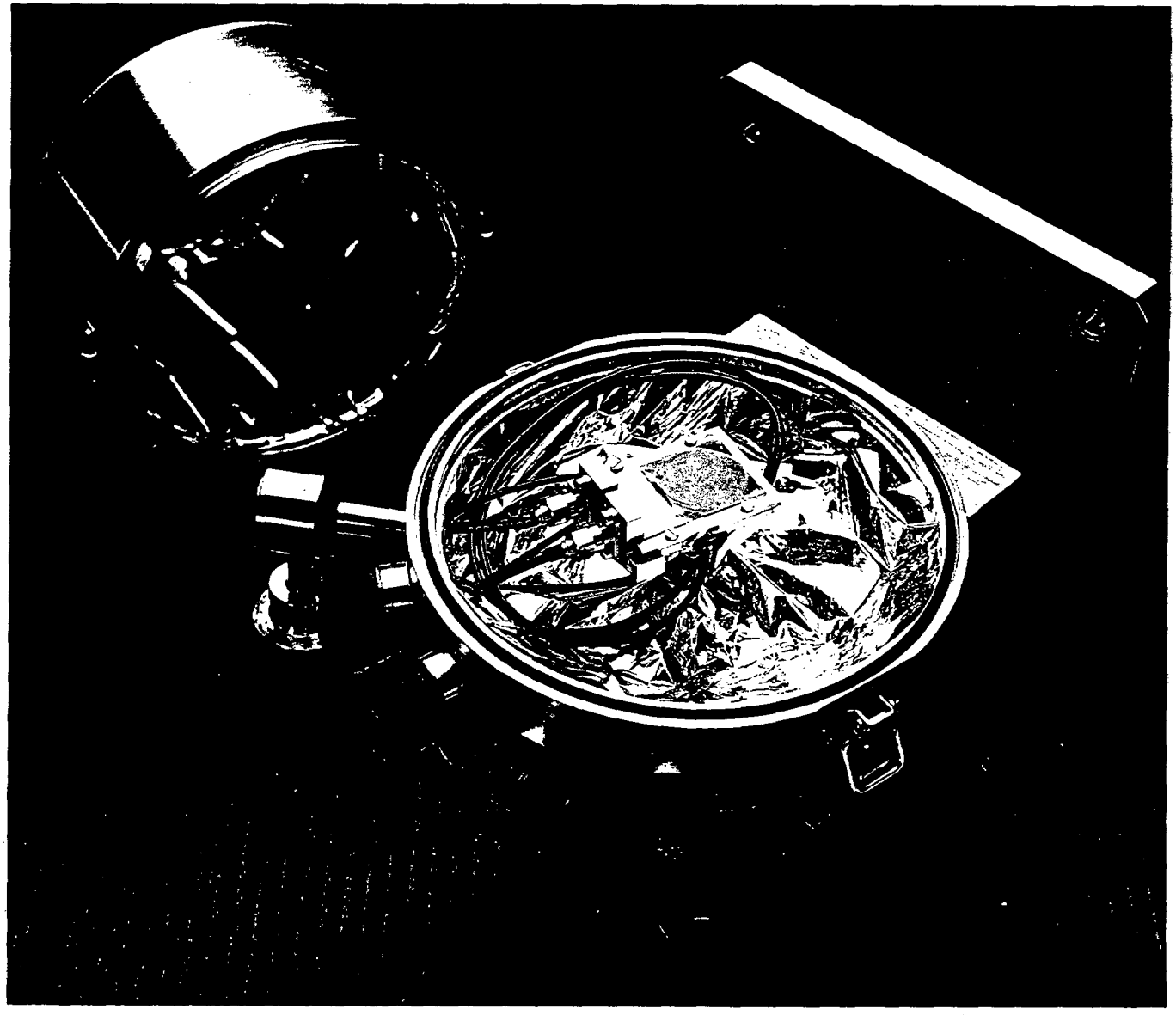

Figure 4: Stirling cryocooler/delay line demonstrator.

due to thermal radiation and conduction. Itwas estimated that with careful minimisation of the heat load to the sample enclosure, the 500mW design would be just sufficient for our needs. This mechanism is 'based on repeated isothermal expansion and compression of the helium charge gas through the regenerator.

A vacuum enclosure and sample support platform was designed and built Oxford Instruments to fit around the Stirling cryocooler. The delay line samples are mounted in the ERA substrate carrier which is attached to a copper back plate. The completed unit is shown open in figure 4.

4 RESULTS

4.1 HTS Powder and Target Preparation

Processing parameters were optimised and dense (96% of theoretical density), pure targets were prepared from commercial YBCO powder. Large diameter, 2 and 4 in targets were fabricated from this powder

YBCO and ReBCO powders (Re: Nd, Eu. Gd, Dy, Ho, Er) were produced by 'solid state reactions and the calcination conditions were thoroughly investigate,.

Dense, high purity, stoichiometric targets, with optimum superconductive properties (exhibiting Tc above 90 K) were prepared from these powders. Sintering and annealing conditions were optimised and the effect of processing parameters on the fins" properties of targets was determined.

4 . 2 Epitaxial HTS Film Deposition

4.2.1 NKT PLD Facility

NKT has during the project upscaled the maximum film size for laser ablation *from* **10x1O mm to 2" diameter. This** step has a great technological importance because it makes it possible for NKT to produce films for superconducting applications requiring large area films e.g. passive microwave components, pickup coils for SQUIDS, shielding, etc. Moreover preparation of large area films greatly reduces production costs for small area films.

NKT has developed depositions on $LaAlO_3$, MgO and Al_2O_3 as an alternative to $SrTiO_3$. This is very important for high frequency applications, because $SrTiO_3$ has very high dielectric losses.

NKT has improved its surface quality significantly. The surface quality is important for making films with low surface resistance and for the reproducibility of sub-micron lithography. The reproducibility of submicron lithography has to be very high to make Josephson junction array based components.

4.2.2 **University of Linz PLD Facility**

The properties of $REBaSrCu_2O_x$ (REBSCO) ceramics were studied for RE = La, Pr, Nd, Sm, Eu, Gd, Dy, Y, Ho, Er, Tm, and Lu. In contrast to RE-123, the critical temperature varies strongly with the ionic radius. On increasing ionic radius the structure changes from orthorhombic to tetragonal. In the transition regime polymorphic compounds can be formed with RE = Gd, Dy. With small RE *ions, the* formation of impurity phases cannot be avoided. The Lu compound cannot be synthesised at all by standard solid-state reactions.

With Y-123 thin films critical temperatures of T_{c0} = 88-89 K and critical current densities of j_c(77 K) = 2-3.106 A/cm^2, both determined by transport measurements, could be reproducibly obtained. Up to 27 lines can be fabricated and measured separately' on a 10 x 10 mm^2 sample. Optimum films yield a standard deviation of the values of j_c(77 K) measured on one film of 10-20' %. We showed that the substrate temperature, which is necessary to deposit high-quality thin films, can be as low as 630"C, when N_2O is employed instead of oxygen. The film properties are deteriorated by photodissociation of N_2O with an excimer laser beam parallel. "to the target, because thereby NO is produced [Schwab 199 la, 1992a].

Lu-123 ceramics contain large amounts of a second phase. Due to the high non-equilibrium nature of the PLD process, it is possible to synthesise phase pure films from this compound. The characteristic properties achieved were T_{c0} = 90 K, 85 K and 84 K for $SrTiO_3$, MgO and $LaAlO_3$ substrates, respectively. The critical current density jc(83 K) exceeded 10^6 A/cm^2 with $SrTiO_3$ substrates [Schwab et al. 1992b].

The optimum preparation conditions for Tm-123 films were found to be similar to those used with Y-123. The same holds for the superconducting properties, film morphology and structure [Stangl et al. 1994a]. With this compound, the development of the surface features with increasing substrate temperature Ts was studied. Optimum superconducting properties correspond to a film surface which is smooth except of outgrowths.

With Gd-123 targets critical temperatures up to T_{c0} = 93.5 K and critical current densities up to j_c(77 K) = 3.5.106 A/cm^2 have been obtained.

At ambient pressure, $YBa_{2-x}Sr_xCu_3O_y$ can only be synthesised by solid-phase reactions up to concentrations x = 1.2. We have demonstrated that with PLD it is possible to fabricate single phase oriented films with x up to 1.8. The length of the c-axis decreases about linearly with increasing x. Correspondingly, the critical temperature decreases e.g. with (100) $SrTiO_3$ substrates T_{c0} = 80 K was found for x = 1. The critical current density was j_c(60 K) 10^6 A/cm* [Schwab et al. 1993a].

LuBaSrCu3Ox cannot be synthesised at all by standard solid-state reactions. We succeeded in preparing single-phase films by PLD at relatively low substrate temperatures of about 600°C [Schwab et al. 1992 b]. To oxidise, the films completely at these low temperatures, N_20 was employed. With such preparation conditions the film surface was granular on a sub-micrometer

scale and the number of particulate was smaller. The superconducting properties were T_{c0} = 54 K and j_c(40 K) 10^5 A / c m * .

The optimum preparation conditions for films $TmBaSrCu_3O_x$ and the surface corresponding to optimum superconducting properties are strikingly similar to Tin-I 21 films [Stangl et al. 1995a]. The maximum value for T_{c0} achieved with (100) MgO, 72.7 K, slightly exceeds the value for ceramics. "The maximum critical current density was j_c(57 K) 10^6 A/cm^2. The optimum window of substrate temperatures seems to be smaller than for Tm- 123 films. Changing the temperature resulted in the formation of sub-mm outgrowths or platelets. The differences in the shape of the Jc(T) dependencies for TBSCO and Tm- 123 films can be discussed in terms of a wider distribution of critical temperatures in TBSCO films. This, in turn can be attributed to a more, inhomogeneous spatial di st ribut ion of oxygen.

The maximum T_{c0} of single phase $GdBaSrCu_3O_{x-}$ thin films achieved was 77 K This is lower than the reproducible level obtained with ceramics, T_{c0}= 80 K, as in the case of Y- I 23. The surface exhibits mainly nearly spherical, droplet-like features, in contrast to TBSCO films. With standard deposition conditions Tc_0 was decreased, if long dwell times at low temperatures and slow cooling rates were employed. This degradation of T_{c0} could be avoided by reducing the target-substrate distance. Maximum critical current densities were around j_c(57 K) = 10^6 A/cm^2.

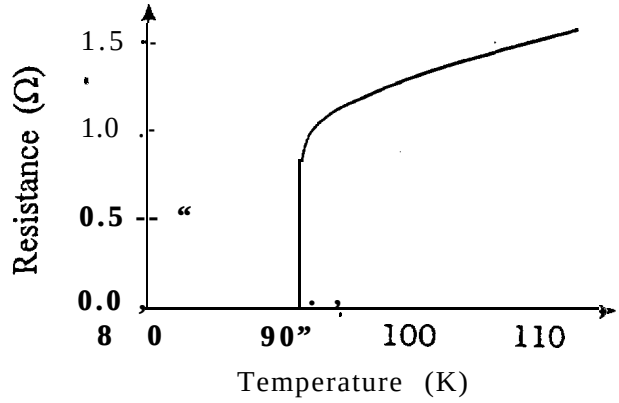

Figure 4: Resistance versus temperature for a YBCO sample produced using the BaF_2 process with ex-situ annealing. The thickness was 500 nm, TCo is 91.6 K and the transition width 0.5 K. The critical current density Jc at 77 K was 2,1x105 A/cm2. The Jc value could increased by decreasing the film thickness.

In order to achieve Bi-22 12-films with T_{c0} = 80 K high deposition temperatures -T, = 800°C - and high deposition pressures - $p(O_2)$ = 3 mbar - were employed. The films were exposed to these conditions also for about 1.5 hours immediately after the deposition. Maximum critical

current densities - j_c(30 K) = 2.1'06 A/cm* - were obtained for T, = 780°C, corresponding to a T_{c0} of 68 K.

4.2.3 Technical **University of Denmark MBE Facility**

During the project we have produced high quality superconducting $YBaCu_3O_{7-x}$ **thin films on** single sided and double sided 10x 10 mm^2 **and** 25x25 mm^2 $LaAlO_3$ substrates. **Also,** we have demonstrated that the process can be extended to include 2 inch' substrates. Figure 4 shows a resistance versus temperature curve for a sample.

From the experiments on double layer depositions and reduced annealing times, we fabricated a double layer film with a thickness of 300 nm with two deposition and two annealing processes, where the annealing time in the high annealing temperature step was reduced to 15 minutes. The results were good in that Jc was increased in comparison with a single layer 300 nrn film (from 2.2x 10^6 A/cm* to 4. lx 10^6 A/cm^2). The surface morphology of the double layer 300 nm film was comparable to that of the single layer 300 nm film.

Double sided l "xl" samples with 300 and 500 nm YBCO superconductor on $LaAlO_3$ **substrates have been made.** T_c**s were just below 90 K** for each side of the 300 nm films, while for the 500 nm films, T_cs were above 91 K. The 77 K microwave surface resistance was measured at 18 GHz (300 nm) and 24.4 GHz (500 nm). Scaling the results to 10 GHz (using the f^2 law) and taking into account the finite thickness effects (correction made by dividing R_s by coth (d/λ), where d" is the thickness and $\lambda(T)$ is the magnetic penetration depth) we found R_s(300 nm @ 77 K)= 0,85 mΩ and Rs(500 nm @ 77 K)= 0.72 mΩ .

4.2.4 **Demokritos Sputter Facility**

Good results have been obtained with *RF sputtering* from a *stoichiometric powder target in an* on axis configuration. The benefits of sputtering from a powder target is that dense YBCO disks of 5 cm diameter are not required, it is easy to change the stoichiometry of the target and overheating of dense targets during sputtering is prevented. The disadvantage is that frequent replacement of the powder is required in order to have films with good properties. Thee films , had very rough surfaces and low J_c and so were worthless for technological applications.

In order to improve the film properties we used a new technique which is based on the *insertion of a conducting (copper) disk* between the target and the substrate and RF excitation of the source. " In this way it was able to use dense targets too with very good results. It is considered that the energetic particles responsible for resputtering are prevented from reaching the growing film because of the disk. Films with excellent surface smoothness, T_c =85 K and J_c(77 K) > 1x 10^6 A/cm* was made on $LaAlO_3$ substrate. However this technique does not permit to cover more than 1 x I cm^2 substrates.

In order to sputter at *higher* pressures we turn to *DC excitation* of the sputter source with dense , and fully oxygenated (and so conductive) YBCO targets. In this way we have succeeded to deposit films with T_c =90 K and JC(7'7K)='4 x 10^6 A/cm^2 Up to l"xl'.

	ON-AXIS RF	ON-AXIS-RF DISK	ON-AXIS
SUBSTRATE T (“c)	680(MgO) 720($LaAlO_3$)	680(MgO) 720($LaAlO_3$)	750(LaAl
TOTAL PRESSURE	200mtorr	2 0 0	450
OXYGEN PRESSURE	15mtorr	15	50
POWER (WATT)	50	50	70

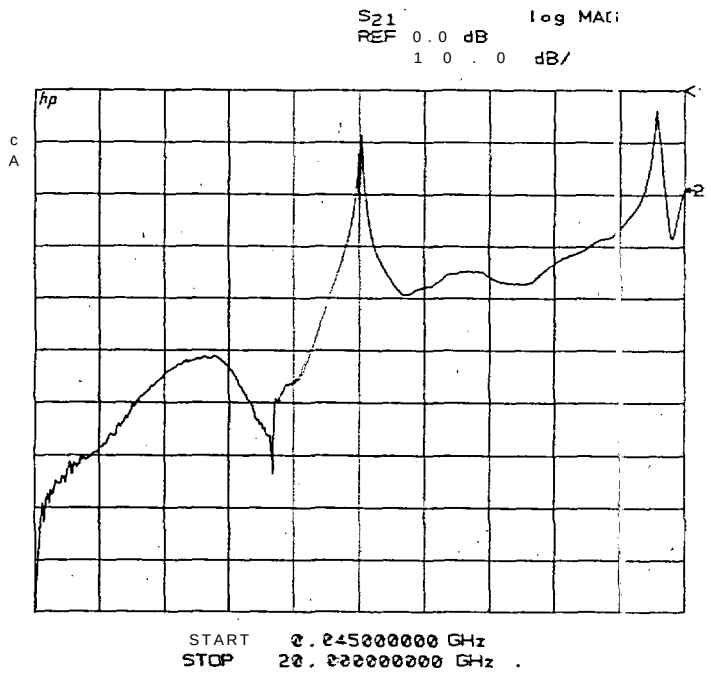

Figure 5: Transmission of an HTS ring resonator as a function of frequency.

4.3 Device Fabrication and Testing

Figure 5 shows the transmission of a YBCO ring resonator **as a function of frequency. A sharp transmission peak** at the fundamental resonance can be seen. This occurs approximately when the ring circumference is a whole number of wavelengths long. The width of this peak is used to determine the Q of the structure. Enlargement of the peak shows that in fact the peak was split in two. This effect is caused by variations in the substrate permittivity due to twinning. It can be avoided by using line-resonator structures, although these were less suitable for characterising film losses because they have higher radiation losses than ring resonators. Figure 6 shows the measured performance of the filter. The insertion loss is ldb less than the gold version and exhibits the characteristic steep-sided response (at the expense of pass-band ripple) of the Chebychev design. Figure 7 shows the performance of a coplanar spi -al delay line produced on a

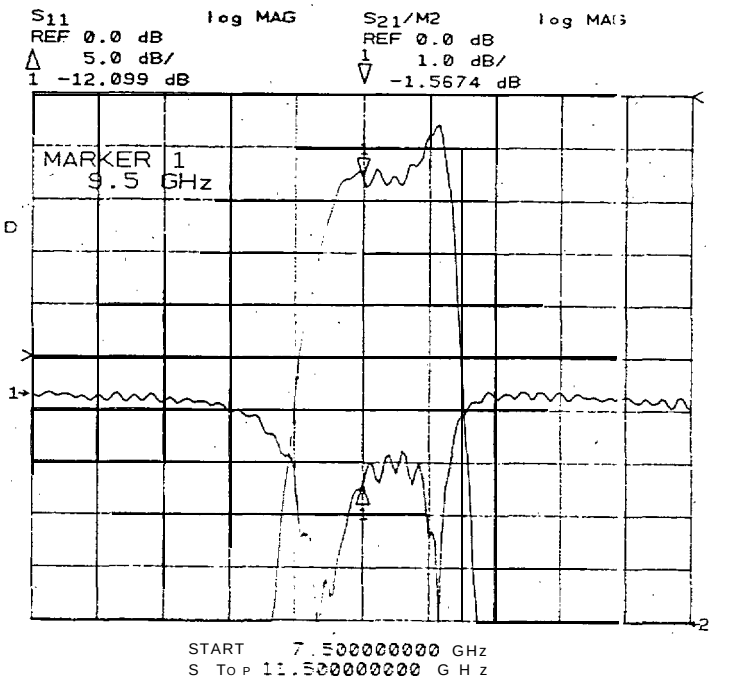

Figure 6: Transmission of three-pole Chebychev filter

1 cm substrate, this gave good performance with the smaller track width giving higher delay (1.9 ns) than a **50 Ω microstrip design on the same substrate area.**

The 1 inch and two inch diameter devices were less successful, showing a periodic structure in their transmission characteristics. Voltage contrast electron microscopy of a two inch device showed a break in the track, caused by contamination of the substrate by a particle, either during growth, transport or patterning. Such faults should be relatively easy to eliminate although high standards are required, unlike normal silicon chip production, delay nines require the whole wafer to be perfect,

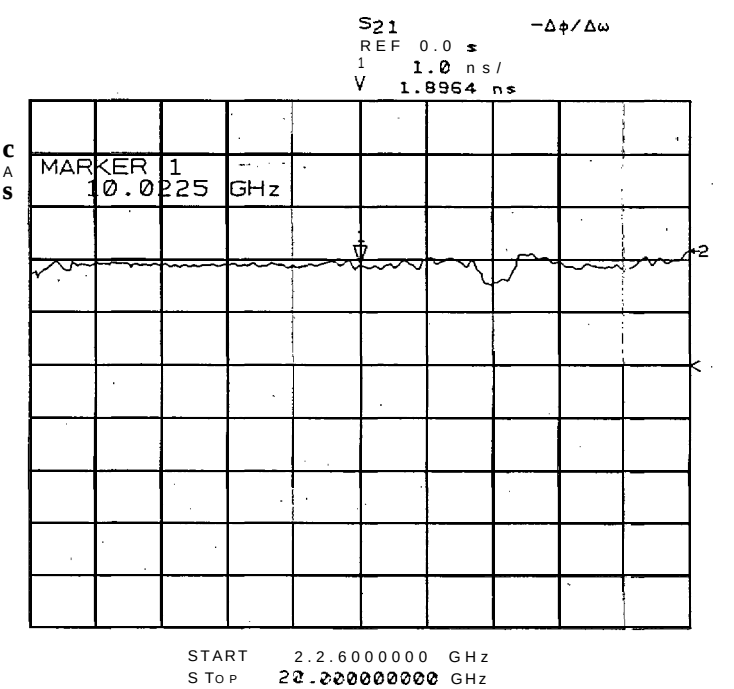

Figure 7: Group delay characteristic of a 10 mm diameter coplanar delay tine

4.4 Cryocooled delay line

This unit successfully achieved cool-down, operating with a 1 inch delay line.

5 CONCLUSIONS

Work by this consortium and by *other* groups have brought HTS microwave technology to the point where, with the important exception. of high power applications, exploitation of the technology now depends on development (rather than research) of components, sub-systems and cryogenic engineering developments. A complete demonstration technology for the fabrication of HTS microwave devices exists and they have much better per formance than conventional microwave components can offer. This is a real achievemer t. However, conventional technologies do not require a cryosystem and therefore if real system benefits are to be realised through application of HTS the advantages have to outweigh the extra complications of employing cryogenic temperatures which must be reduced to a mininum.

6 ACKNOWLEDGEMENTS

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