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**REDUCED NONLINEARITY SUPERCONDUCTING THIN FILMS TO TRANSMIT
AND RECEIVE APPLICATIONS**

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14. ABSTRACT This project is directed towards the development of high performance large-area YBCO films on suitable single crystal substrates through modified TFA-MOD approach for transmit and receive applications. The research has lead to the state-of-the-art of development of high quality YBCO films on single crystal substrate with $T_c(R=0) > 92K$ and $J_c > 5MA/cm^2$. The linearity, surface resistance, and power handling capability of YBCO films developed in this research are greatly improved by the understanding and control of driving force and HF saturation in precursor during the conversion. Mechanism study indicates that layered growth is critical and can be realized by surface modification and nucleation control. A nucleation transition from dendrites to layered structure has been revealed. It has been approved that the modified TFA-MOD process is a very low cost route for YBCO fabrication and can be scaled up for high frequency applications.					
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1.0 SCOPE

This project is directed towards the development of high performance large-area thick YBCO films and microwave devices on suitable single crystal substrates through modified TFA-MOD approach for transmit and receive applications. We propose to continue our success in Phase I to further increase critical current density of J_c to 3-5 MA/cm² and lead to state-of-the-art IMD (intermodulation distortion) by improving linearity, reducing surface resistance and increasing power handling capability of YBCO films. The major focus for UES is to develop high performance YBCO films with J_c greater than 3 MA/cm² at 77K self field by modified TFA-MOD process through coating, pyrolysis, and crystallization. To achieve these goals, layered growth mode is suggested and will be pursued by the control of driving force and HF saturation of precursor. The UES approach would make film fabrication much cheaper, highly reproducible, and easy to scale up. The process will be scaled up to make large area thick YBCO films for fabricating large arrays of microwave filters for the transmit and receive applications for both military and civilian applications

2.0 INTRODUCTION

High quality HTS (high-temperature-supereonductor) thin films, in partieular, the YBa₂Cu₃O_{7-δ} (YBCO, Y123) films, open the new field for electronics. Mierowave frequency filters made with high-temperature-superconductor materials have outstanding performance, accomplishing filter specifications for sharp cutoff and narrow bandwidth that in practical systems cannot be realistically achieved with conventional technology. There are many situations where YBCO filters will solve important interference problems as they have in cellular telephone base stations. Several successful field tests in military systems have also been reported. But they are limited in the power that can be passed through. This limits such filters to receive only applications. However, even in low power receive applications the filters can produce intermodulation distortion (IMD). The origins of the limited power handling and the IMD are the same: the nonlinear nature of the surface impedance. The IMD has obviously not been a problem in the filters that have been used so far but is likely to be in the future when filters and systems that stress the limits of performance are employed. The quality of the HTS film has improved over the years and the experimental evidence indicates that the intrinsic limit of the microwave nonlinearity of the HTS materials has been reached in the best films presently available. The limit is imposed by the d-wave symmetry of the HTS materials and the associated nodes in the energy gap, leading to nonlinearities that are enhanced over those of the older low-temperature s-wave materials. The experimental agreement with theoretial predictions is one of the important facts pointing to the achievement of the intrinsic limit.

3.0 MAJOR TASKS ACCOMPLISHED

We proposed the following tasks for this project. Currently we have all of them.

Task 1: Crystal Selection, Surface Preparation, and CeO₂ Buffer Growth

Task 2: Modified TFA Solution and Thick Precursor Preparation

Task 3: Thicker Film Fabrication

Task 4: High Quality YBCO Growth

Task 5: Investigation of Fine-Tuning of Microwave and RF Properties by Optimization of YBCO Nucleation and Growth

(1) Investigation of the Effect of Humidity on Electrical Properties

(2) Investigation of the Effect of Oxygen Partial Pressure on Electrical Properties

(3) Investigation of the Effect of Low Pressure Conversion

(4) Investigation of the Effect of Conversion Temperature

(5) Investigation of the Effect of Gas Flow

Specifically, for Task 1, we have accomplished crystal selection, Surface Preparation, and CeO₂ Buffer Growth. We use LaAlO₃ single crystal substrates to growth YBCO films. Nevertheless, even though all providers provide single crystal substrates, because of crystallinity, the crystal purity, twins, precision on the orientation, quality of polishing, so on so forth, some of the substrates are not suitable for high quality YBCO growth. After careful comparison, we have selected a provider who can provide high quality LAO wafers with low cost. No much difference was observed for batch to batch. With these high quality LAO wafers we have been able to further prepare the surface by surface cleaning followed by buffer layer growth of CeO₂. We have developed a unique method for the buffer growth by magnetron sputtering. High quality CeO₂ layer was grown successfully and highly reproducible. This CeO₂ buffer has a very good crystallinity, which guaranty the quality of YBCO to carry a critical current density over 4 MA/cm² routinely.

For Task 2, we have developed the modified TFA solution (MTFA) several years back and in this research, we have been modifying this MTFA solution in terms of the way of the solution synthesis and the concentration. With these modifications, the stability of the solution was improved quite a lot and very long shelf life has been reached. We have not see obvious difference on the YBCO quality for solution synthesized few months back compared with only few days of the TFA YBCO precursor solution. By using rotary evaporator, the viscosity and concentration of the MTFA solution can be easily adjusted; for example, the highest concentration of the solution can be as high as 2.5M and can be diluted to any concentration bellow. This brings the advantage of fabricating films with different thickness. We have realized thick films of the about 2.0 μm by coating and burnout three times.

For Task 3, we are working on the thicker film fabrication. We have realized thick films of the about 2.0 μm by coating and burnout three times. Nevertheless, there are two major issues as limiting factors for the high quality YBCO films. One is film cracking during the burnout and the other is J_c degradation. We have progress on film cracking control by adjusting concentration/viscosity and surface status of substrates. Three coats without cracks for the MTFA solution were demonstrated. This process needs to be reconfirmed and made into SOP.

Another issue is the degradation of the thick YBCO films. We have been modifying the conversion process to improve the quality of YBCO films. On the other hand, we are improving the performance of single coating of YBCO films. This is an alternative to increase the power handling capability rather than through thick films.

For Task 4 and Task 5, in the past year we have been realized high quality YBCO films by the control of growth. We have achieved very high critical currently density of $5.15\text{MA}/\text{cm}^2$ at 77K self-field. Figure 1 shows the result we have got in the past year. We developed a process that can engineering the substrate surface to form nanostructure for YBCO nucleation and growth, which improve the critical current dramatically. The performance of the MOD YBCO films has been improved from previously about $2\text{MA}/\text{cm}^2$ to over $5\text{MA}/\text{cm}^2$. We can easily reproduce high currently density about $4\text{MA}/\text{cm}^2$ with this new approach.

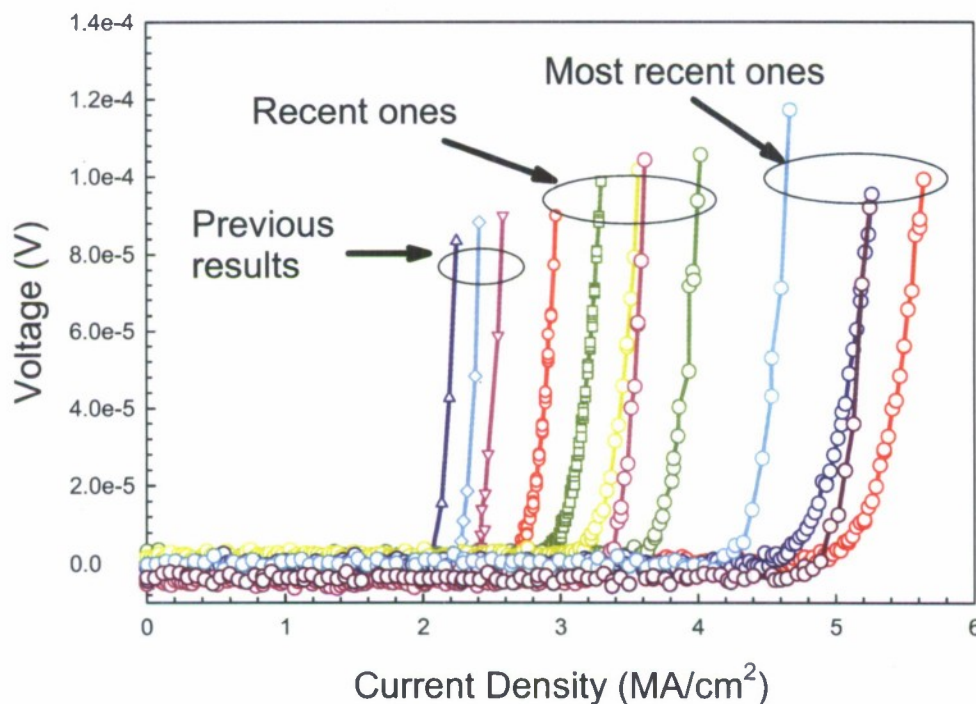


Figure 1. Critical current density of modified TFA-MOD YBCO films at 77K self-field

To achieve high performance YBCO films, major efforts have been taken on nucleation and growth of YBCO films. First, the nucleation transition from dendrite, cell to layered mode as shown in Figure 2 was revealed found. This is a critical step for high critical current density, because large grains and grain boundaries are the source of weak links. By increasing the driving force, we have been able to control GBs and growth rate thus improve I_{cs} and J_{cs} . By adjusting the driving force (temperature, humidity, and oxygen partial pressure), a transition of YBCO nucleation on LAO from dendrite (at very low driving force) to cellular (at intermediate driving force) and to "good epitaxial" (at relative high driving force) can be established. At high growth rate (high driving force), no dendrites and cellular microstructures either at interface or on the top surface of the as grown film were observed indicating nucleation/growth transition might have changed to layered mode.

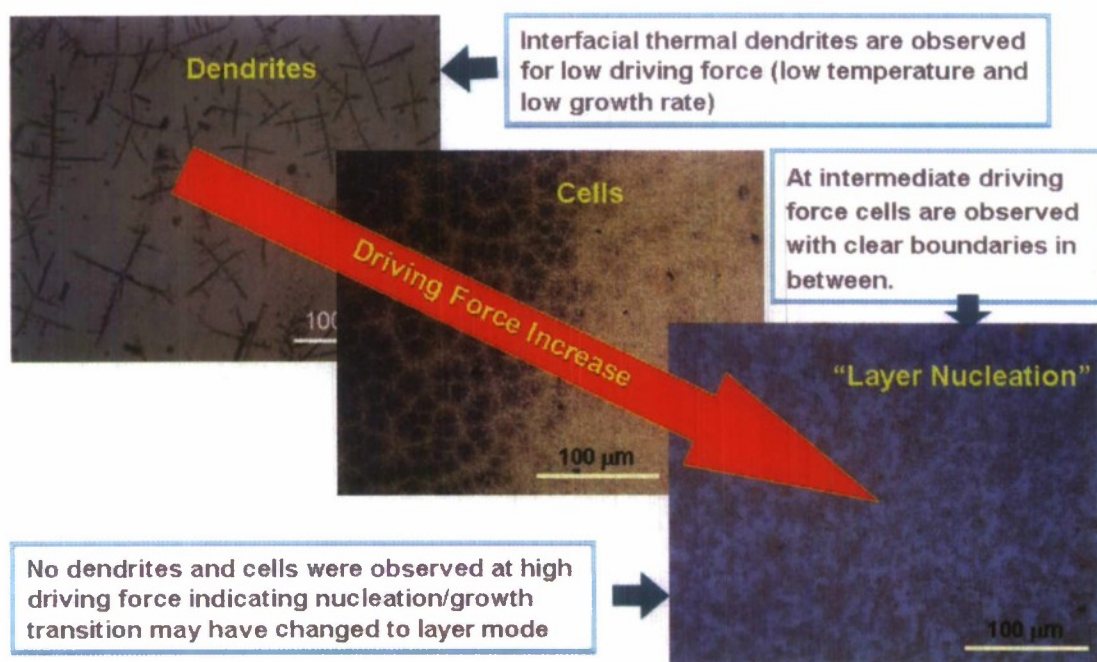


Figure 2. By adjusting the driving force (temperature, humidity, and oxygen partial pressure), a transition of YBCO nucleation on LAO from dendrite (at very low driving force) to cellular (at intermediate driving force) and to "good epitaxial" (at relative high driving force) can be established. At high growth rate (high driving force), no dendrites and cellular microstructures either at interface or on the top surface of the as grown film were observed indicating nucleation/growth transition might have changed to layer mode.

A key step has been taken to improve the surface quality by solution approach as shown in Figure 3. With this step, we have been able to increase J_c from 2~3 MA/cm² to above 5MA/cm².

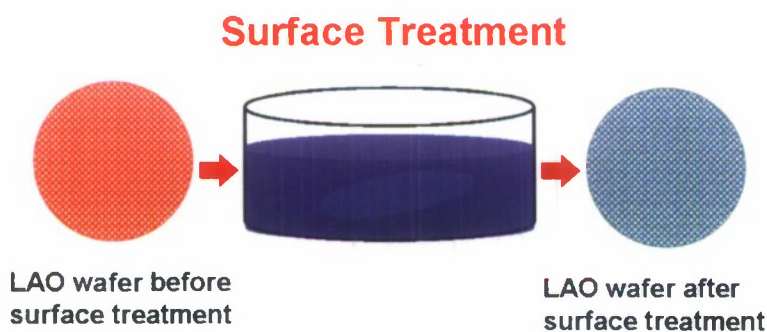


Figure 3. A key step developed for engineering nanostructured surface for YBCO nucleation and growth

With the increase of driving force, critical current density increases monotonously and high J_c of 5 MA/cm² has been measured as shown in Figure 1.

One of the issues is that high self-field J_c does not mean high performance in field. For example as shown in Figure 4 (a), the J_c drops very fast from 5.15MA/cm^2 to 1.7MA/cm^2 at the field of 377 Gauss, even though this is a very small field. The angle dependence shows the same trend of J_c . For the sample with best self-field J_c , $J_c(\perp B)$ is only about 37% of $J_c(\parallel B)$ at 377 gauss, Figure 4 (b).

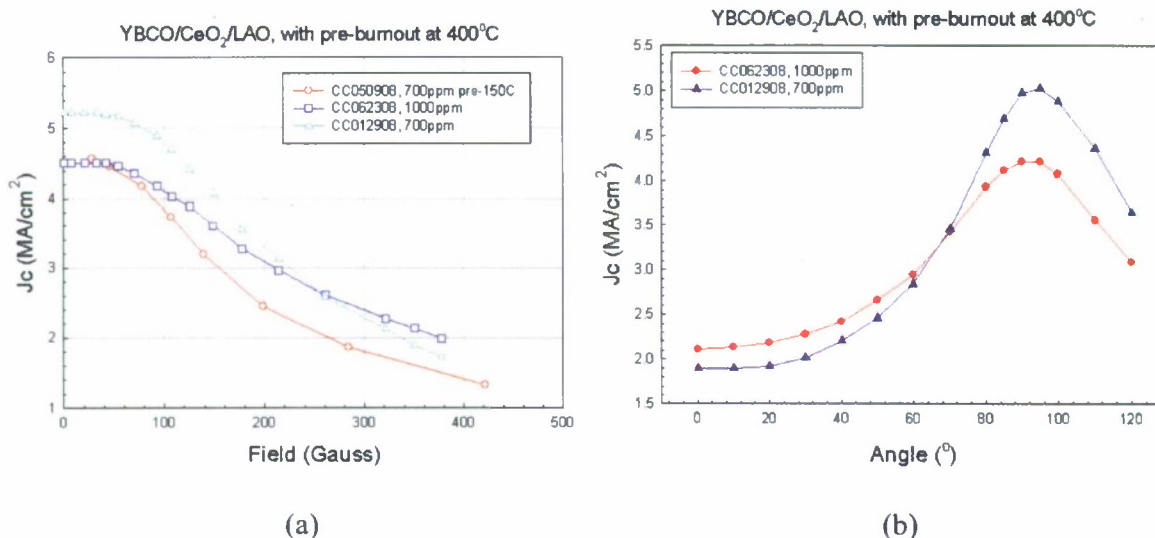


Figure 4. (a) The in field performance of MOD YBCO films by modified TFA-MOD approach and (b) The angle dependence of MOD YBCO films by modified TFA-MOD approach.

We further adjusted the processing conditions and a much better performance has been achieved. With this adjustment, some sacrifice of the self-field J_c was observed, however, the infield J_c was improved from 37% of the best self-field sample to 65%. The absolute value was also improved from 1.7MA/cm^2 to 2.31MA/cm^2 , which is about 35% increase (Figure 5 (a)). The angle dependence is also improved greatly, Figure 5 (b). The difference for the $J_c(\parallel B)$ and $J_c(\perp B)$ is very small and the line is almost flat for all angles. This film is even better than those films deposited by PVD approaches. Figure 6 summarizes the in-field performance of some of the YBCO films prepared in our lab including those by magnetron sputtering on different substrates. Figure 7 summarizes the angle dependence of the samples prepared in our lab including magnetron sputtering YBCO films on different substrates. It should be noticed that, all of the measurements for the angle dependence were performed in a field of 377 Gauss. The in-field and angle dependence improvement was attributed to the pinning effect. The efforts to make better in-field performance are to make microstructure finer. A criterion for better pinnings is to avoid dendrite and coarse nucleation, instead, control very fine microstructure and layered nucleation (or super small grains). Figure 6 and 7 are some of the recent results for samples processed in different conditions.

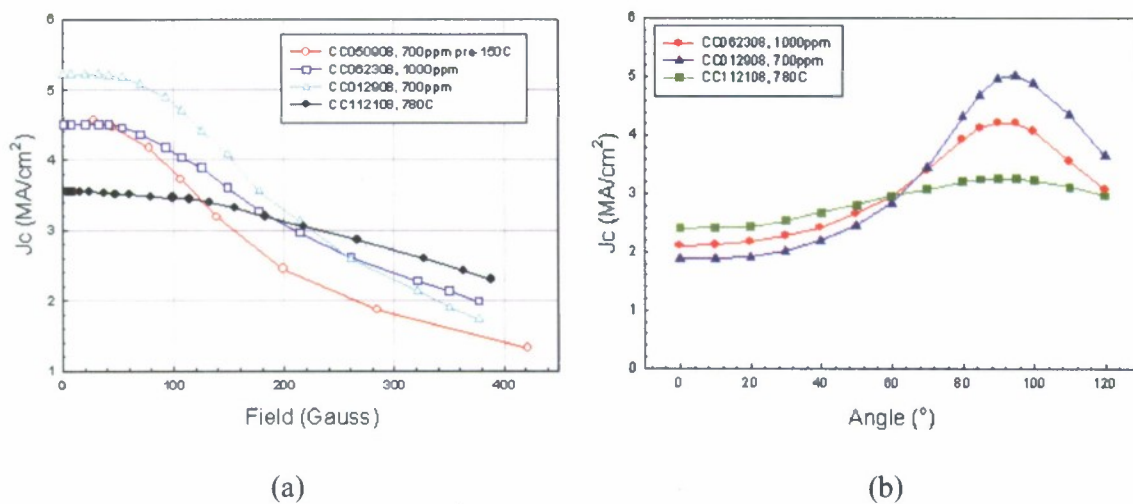


Figure 5. A comparison of MOD YBCO samples with/without strong pinning enhancement. (a) The in field performance of MOD YBCO films by modified TFA-MOD approach and (b) The angle dependence of MOD YBCO films by modified TFA-MOD approach.

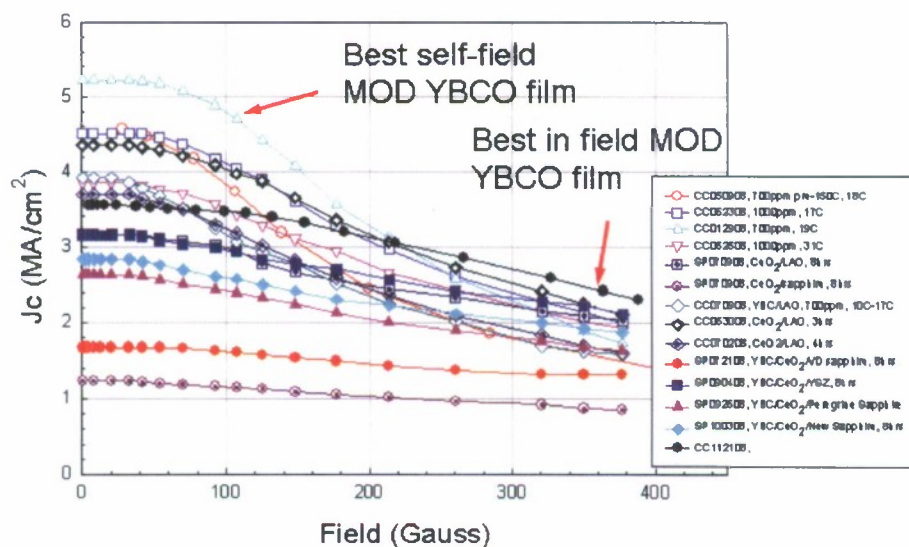


Figure 6. The in field performance of MOD YBCO films by modified TFA-MOD approach under different processing conditions.

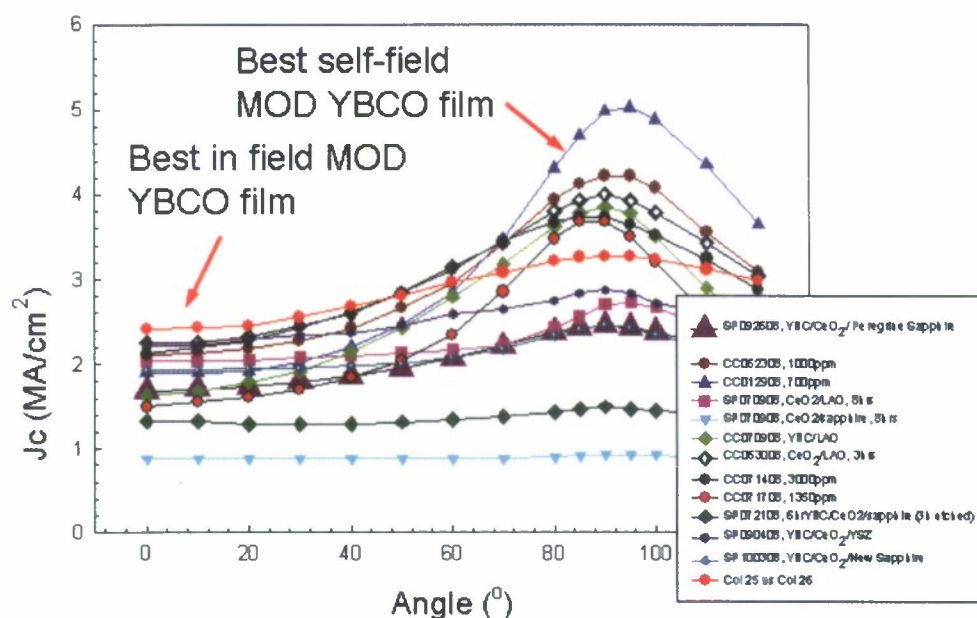


Figure 7. The angle dependence of MOD YBCO films by modified TFA-MOD approach under different processing conditions.

As a result, the improvement of intermodulation distortion (IMD) was observed, as shown in Figure 8. Those samples that show good in field performance are in the processing and have not completed the IMD measurement. Samples with high J_c s (self-field) and good microstructures show the improvement both at low power and high power ranges.

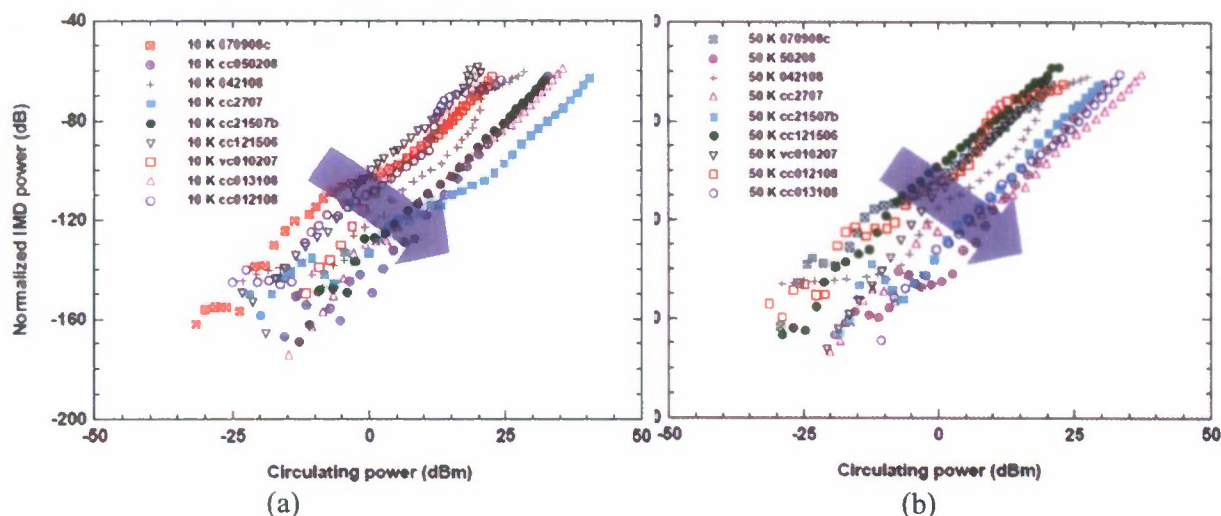


Figure 8. Intermodulation distortion (IMD) measurement showing improvement of IMD by nucleation and microstructure control. (a) Data measured at 10K and (b) measured at 50K.

4.0 SUMMARY

In this research, we have succeeded in accomplishing the proposed goals both on theoretical understanding and on electronic performance of MOD YBCO films. The progress can be enumerated as follows:

- Theoretical understanding of the nucleation and growth of YBCO films, a mechanism of nucleation transition from dendrites to cells to fine grains and to layered is proposed. One journal paper is accepted for publication.
- High critical current density of $5.15\text{MA}/\text{cm}^2$ at 77K self-field has been reached.
- Much better pinning effect has been realized in terms of in-field performance and angle dependence.
- We have attained the best IMD of our MOD YBCO films that we have measured so far at the low power end.
- YBCO processing is highly reproducible and low cost. We can provide high quality YBCO films on 2" LAO wafers.

5.0 FUTURE WORK

The current research is successful while it is worth well for even better performance on thick films. YBCO films through MOD approach are one of the most cost effective methods for electronics applications. Current results show that further control nucleation and in particular, microstructure should be the major focus for the future research. The pinning effect that improves the in field performance can be a big factor for better IMD performance. We will further investigation how to make the microstructure finer with better pinning effect. This will involve a precise processing condition control for better nucleation and growth. In addition, strip line resonator design will be carried out in future research.



HYBRID APPROACH FOR MULTI-SCALE MODELING OF RADIATION TRANSFER IN THREE-DIMENSIONAL NON-GRAY MEDIA

STTR Phase I Final Report

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