

## MIT Open Access Articles

*Intermodulation Distortion in Epitaxial Y-Ba-Cu-O Thick Films and Multilayers*

The MIT Faculty has made this article openly available. **Please share** how this access benefits you. Your story matters.

**Citation:** Ho Won Jang et al. "Intermodulation Distortion in Epitaxial Y-Ba-Cu-O Thick Films and Multilayers." Applied Superconductivity, IEEE Transactions on 19.3 (2009): 2855-2858. © 2009 IEEE

**As Published:** <http://dx.doi.org/10.1109/TASC.2009.2019668>

**Publisher:** Institute of Electrical and Electronics Engineers

**Persistent URL:** <http://hdl.handle.net/1721.1/52619>

**Version:** Final published version: final published article, as it appeared in a journal, conference proceedings, or other formally published context

**Terms of Use:** Article is made available in accordance with the publisher's policy and may be subject to US copyright law. Please refer to the publisher's site for terms of use.



# Intermodulation Distortion in Epitaxial Y-Ba-Cu-O Thick Films and Multilayers

Ho Won Jang, Kyoung-Jin Choi, Chad M. Folkman, Daniel E. Oates, and Chang-Beom Eom

**Abstract**—We report intermodulation distortion (IMD) in high-quality epitaxial  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) thick films and multilayers prepared by pulsed-laser deposition (PLD) using multiple targets. Two sets of films were prepared: single-layer and multilayer YBCO films in which a 20-nm-thick  $\text{CeO}_2$  inter layer is inserted between every 100-nm-thick YBCO layer. With increasing YBCO thickness from 200 nm to 1200 nm, single-layer films exhibited improvement of IMD, whereas multilayer films showed degradation of IMD. Similarly, YBCO crystalline quality in single-layer films was improved with increasing the thickness, while that in multi-layers films degraded. These results suggest that there is a strong correlation between crystalline quality and IMD for YBCO films. The comparison of experimental data with the theory of IMD suggests that improving crystalline quality is essential for YBCO films with thickness greater than  $1\ \mu\text{m}$  to achieve an intrinsic nonlinear behavior. PLD using multiple targets is a very useful method to grow thick YBCO films with high crystalline quality and low IMD.

**Index Terms**—Intermodulation distortion, nonlinearity, pulsed-laser deposition,  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ .

## I. INTRODUCTION

NONLINEARITIES at microwave frequencies in the cuprate high-temperature superconductors (HTS) remain a subject of great interest, both because of the basic physics of the materials and because of the implications for practical devices such as filters. The linear (low-power) surface impedance is low enough to support very demanding microwave-frequency filter applications. Thus, a recent focus has been the study of the nonlinear effects. The most important manifestation of the nonlinearity is intermodulation distortion (IMD), which can be a limiting factor in HTS filter performance. It is therefore desirable to seek means to reduce the IMD resulting from the nonlinear surface impedance. Recently, a new rigorous theory of IMD in HTS has been formulated and shown to agree well with experiments [1]–[5]. The theory indicates that the nonlinearity in high-quality films results from pair breaking by the microwave current and is intrinsic to the superconductor. Extrinsic causes such as defects and weak links can lead to IMD as well, but can be minimized in high-quality films. The intrinsic

effect is known as the nonlinear Meissner effect [5] and is enhanced in the d-wave cuprates because the nodes in the order parameter lead to easier pair breaking. The agreement between theory and experiment in high-quality YBCO films implies that the IMD is due to this intrinsic cause and improvements in film quality will not further improve the IMD. The theory, however, does predict that the IMD should decrease at least as fast as  $1/t^4$  due in part to the decreased current density in the thicker film and due to the application of nonlocal electrodynamics as required by the theory. Thus, this research seeks to lower the IMD by growing films with thickness greater than  $1\ \mu\text{m}$  while maintaining high quality, which has been a challenge because film quality generally degrades as the film is grown thicker. We have investigated IMD as a function of film thickness for both single-layer and multilayer films as discussed below.

## II. EXPERIMENT

### A. Film Growth and Characterization

We have grown epitaxial  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) films on  $2^\circ$  miscut (001)  $(\text{LaAlO}_3)_{0.3}(\text{Sr}_2\text{AlTaO}_6)_{0.7}$  (LSAT) substrates with an epitaxial 20-nm-thick  $\text{CeO}_2$  buffer layer by pulsed-laser deposition (PLD) at  $760^\circ\text{C}$  in 350 mbar  $\text{O}_2$ . The use of the  $\text{CeO}_2$  buffer layer significantly increased the critical current density due to enhanced flux pinning from a high density of antiphase boundaries, stacking faults, and edge dislocations [6]. Two sets of films were grown. One set is single-layer YBCO films on  $\text{CeO}_2/\text{LSAT}$ , and the other set is multilayer YBCO films, in which a 20-nm-thick  $\text{CeO}_2$  interlayer is inserted between every 100-nm-thick YBCO layer. The schematics of both set are shown in Fig. 1. The total YBCO thickness of the films varied from 200 nm to 1200 nm. In order to prevent the degradation of film quality with increasing the film thickness, multiple YBCO targets have been used. The details of the process will be discussed elsewhere [7]. Crystallinity and in-plane alignment of the YBCO films were examined by high-resolution four-circle x-ray diffraction. The surface of the films was imaged by an atomic force microscopy. The resistivity and critical current density ( $J_c$ ) at various temperatures were measured by standard four-probe method in a bridge geometry patterned by photolithography. For the YBCO films grown in this manner, the transition temperature ( $R = 0$ ) and  $J_c$  were typically measured to be  $90 \pm 1\ \text{K}$  and  $2\text{--}6\ \text{MA}/\text{cm}^2$ , respectively.

### B. IMD Measurements and Simulation

The IMD measurements were carried out on stripline resonators fabricated from films deposited on  $1\ \text{cm} \times 1\ \text{cm}$  LSAT substrates. The films were patterned using standard photolithography and wet etching. After patterning, the etched striplines

Manuscript received August 21, 2008. First published June 30, 2009; current version published July 15, 2009. This work was supported by the Air Force Office of Scientific Research under Grant FA9550-06-1-0415 at University of Wisconsin and FA8721-05-C-0002 at MIT Lincoln Laboratory.

H. W. Jang, K. J. Choi, C. M. Folkman, and C. B. Eom are with the Department of Materials Science and Engineering, University of Wisconsin-Madison, Madison, WI 53706, USA (e-mail: hwjang@wisc.edu; kjchoi@kistmail.kist.re.kr; folkman@wisc.edu; eom@engr.wisc.edu).

D. E. Oates is with MIT Lincoln Laboratory, MA 02420, USA (e-mail: oates@ll.mit.edu).

Digital Object Identifier 10.1109/TASC.2009.2019668

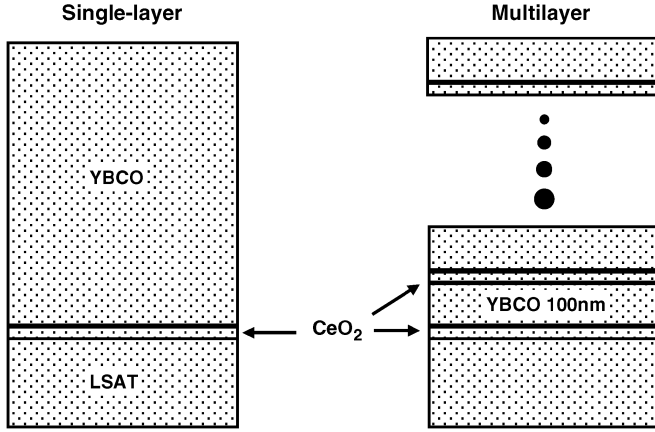


Fig. 1. Schematic of single-layer and multilayer YBCO films. The thickness of the  $\text{CeO}_2$  buffer layer and interlayer is 20 nm.

were assembled with YBCO ground planes to form stripline resonators. The properties of the patterned line dominate the performance of the resonator because the current density is approximately a factor 100 higher in the line than in the ground plane. The linewidth is  $150 \mu\text{m}$ . The line impedance is  $Z_0 = 33$ .

The resonators were measured by a technique that has been described previously [8] in which the quality factor  $Q$  and resonant frequency  $f_0$  of the resonator are measured as a function of the microwave power at temperatures between 5 K and  $T_c$ . The measurements were carried out at the fundamental frequency of 1.5 GHz. The third-order IMD was measured in the usual way, in which two closely spaced tones of equal power at frequencies  $f_1$  and  $f_2$  are combined and applied to the resonator. The frequencies are centered about the resonant frequency with a tone separation of approximately  $1/32$  of the low-power 3-dB bandwidth. The frequencies of the tones were adjusted at each power level and temperature to maintain the same relationship to the bandwidth and resonant frequency. The power  $P_{\text{IMD}}$  of the third-order mixing products at frequencies  $2f_1 - f_2$  and  $2f_2 - f_1$  is then measured in a spectrum analyser as a function of the input power to the resonator. For the analysis of the data, the measured  $P_{\text{IMD}}$  is converted to a normalized IMD power  $P_{\text{NORM}}$ , which removes the dependence of the IMD on resonator unloaded  $Q$ -value and insertion loss [9].

$$P_{\text{NORM}} = P_{\text{IMD}} / r_v (1 - r_v) Q_c. \quad (1)$$

In (1),  $r_v$  denotes the voltage insertion ratio which is related to the insertion loss  $IL$  in dB by

$$r_v = 10^{-\pi/20}, \quad (2)$$

and  $Q_c$  is the unloaded  $Q$  of the resonator. In addition, the input power  $P$  is converted to the circulating power  $P_{\text{circ}}$  associated with the standing wave in the resonator at resonance according to the expression

$$P_{\text{circ}} = 4Q_l r_v P / \pi, \quad (3)$$

In (3)  $Q_l$  denotes the loaded  $Q$  value. The microwave current is related to the circulating power by

$$P_{\text{circ}} = Z_0 I^2. \quad (4)$$

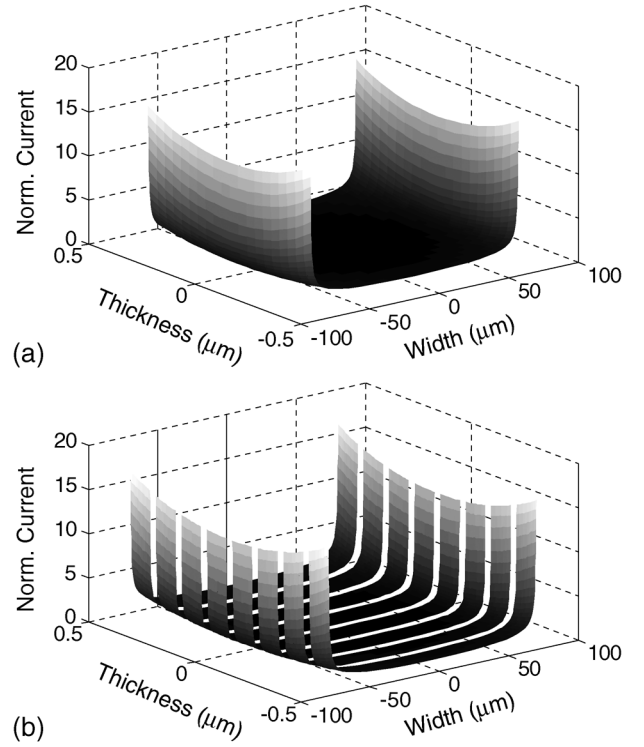


Fig. 2. (a) Current distribution in a stripline employing a single-layer film 800-nm thick (b) current distribution in a stripline employing an eight-layer film each 100 nm with a 20 nm layer between them. For both calculations the strip width is  $150 \mu\text{m}$  and the penetration depth is assumed to be 300 nm.

The data are then plotted as normalized IMD power  $P_{\text{NORM}}$  vs.  $P_{\text{circ}}$ .

The current distribution in the stripline is of great importance when analysing the measured data. Because of the highly nonuniform distribution, changes due to geometry can alter the characteristic impedance of the stripline and calculations of surface impedance from the measured data. In addition, alteration of the current distribution renders the comparison of IMD values difficult because the IMD depends strongly on the current distribution. It is expected however that a 20 nm buffer layer should change the current distribution only minimally, because this value of buffer layer thickness is smaller than a penetration depth and much smaller than the electromagnetic wavelength. In order to assess the effects of the multilayers, the current distribution was calculated for a film composed of eight 100 nm layers with a 20 nm interlayer between them. For the calculation it is assumed that the permittivity of the buffer layer was the same as the substrates. Fig. 2 shows the results of the calculation [10] for the single layer (Fig. 2(a)) and for the eight-layer case (Fig. 2(b)). The results show no significant alteration of the current distribution save for the lack of current in the buffer layer. This justifies treating the resonators made with the multilayer films in the same manner as the single-layer films and the direct comparison of the IMD.

### III. RESULTS AND DISCUSSIONS

#### A. Crystallinity

Fig. 3 shows the full-width half-maximum (FHHM) value of the x-ray rocking curve of 005 YBCO peak as a function

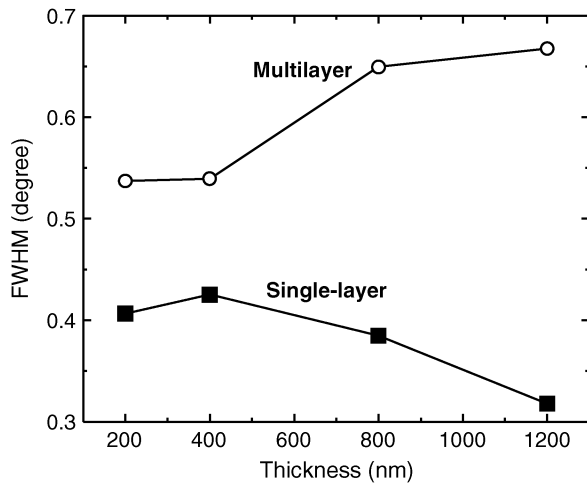


Fig. 3. FWHM of 005 YBCO rocking curve as a function of film thickness for single-layer and multilayer YBCO films.

of total YBCO thickness for both single-layer and multilayer films. As the thickness increases, FWHM of single-layer films is decreasing, while the multilayer films show degradation in FWHM. The enhancement of FWHM in the single-layer films with increasing the thickness is consistent with our observation for YBCO-coated conductors [11]. Thus, it is expected that the insertion of the  $\text{CeO}_2$  layers between YBCO degraded the crystallinity of YBCO in the multilayer films. It is noteworthy that our PLD system using multiple targets allowed us to make the high-quality 1200-nm-thick single-layer film without structural degradation. During PLD, the laser-ablated region in an YBCO target gets deeper and non-stoichiometric with increasing the deposition time. Thus, the crystalline quality of YBCO films usually degrades over the thickness of 400 nm. However, the use of multiple targets could prevent the degradation in film quality, resulting in the fabrication of the high-quality single-layer 1200-nm-thick film.

### B. IMD

Fig. 4 shows typical results of the measurements of normalized IMD as a function of circulating power at a temperature of 50 K for single-layer films with three different thicknesses, 200, 400, and 1200 nm. A complete set of temperatures from 5 to 80 K was measured for each film. For the sake of clarity only the 50 K results are plotted here. As is observed in many such measurements, the curves show different slopes on this double logarithmic plot. These complexities are only partly understood [7], but these data show a slope between 2 and 3 for the majority of the power range. As can be seen in the plot the IMD in the thickest film is lower than in the thinner films.

Fig. 5 shows the normalized IMD as a function of circulating power for the series of films with 100-nm layers of YBCO and 20-nm interlayers between them. The total YBCO thicknesses are 200, 400, and 1200 nm. This plot has the same dimensions as that of Fig. 4. Note that the IMD values are higher for these samples than for the single-layer samples and that the lowest IMD is measured with the thinner films. This is a strong indication that the film quality is evolving differently from the single-layer films.

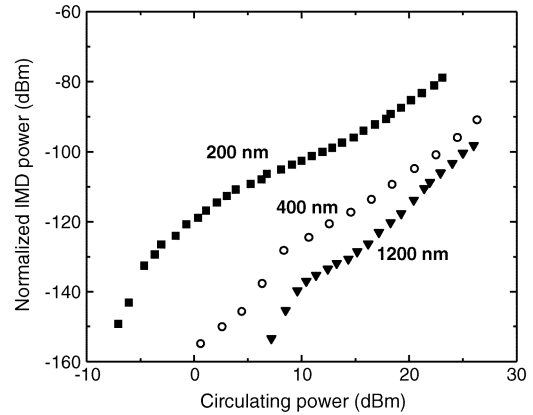


Fig. 4. Normalized IMD as a function of circulating power for single-layer YBCO films.

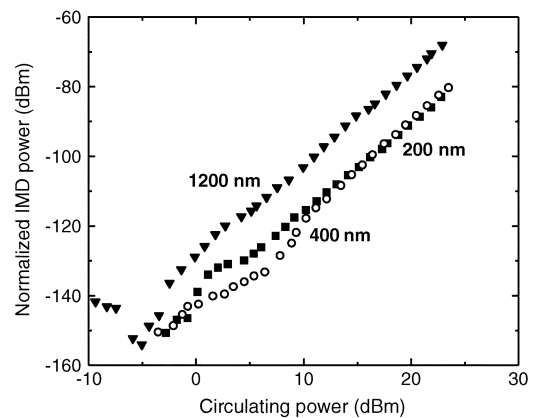


Fig. 5. Normalized IMD as a function of circulating power for multilayer YBCO films.

It has been observed in previous measurements that the IMD increases when the film quality is lowered and the lowest IMD have been measured in high-quality epitaxial films [9]. The results presented here are a quantitative study of IMD as a function of film quality. As discussed in [8], [9], the intrinsic IMD results from pair breaking by the microwave current due to the nodes in the superconducting energy gap in d-wave YBCO and the other cuprate HTS. Thus, pair breaking can occur even at currents orders of magnitude lower than the  $J_C$ . Structural defects may also lead to increased pair breaking by locally lowering the gap. Thus, defects that enhance the critical current may actually degrade the IMD performance. Structural defects may also increase the IMD by acting as Josephson-junction weak links [12]. The nonlinear resistance and inductance of the weak link are responsible for the IMD.

Previous studies have not shown a strong correlation between critical current and IMD. This is to be understood in the observation that IMD occurs at currents that are orders of magnitude lower than the critical current. Thus pinning centers that help to produce high critical currents are not likely to lower the IMD.

### C. Comparison With Theory

Fig. 6 shows the normalized IMD vs temperature for the 1200-nm-thick single-layer film. The data shows the characteristic increase in IMD at low temperature resulting from intrinsic origins in d-wave superconductors as discussed in [1], [2], [3],

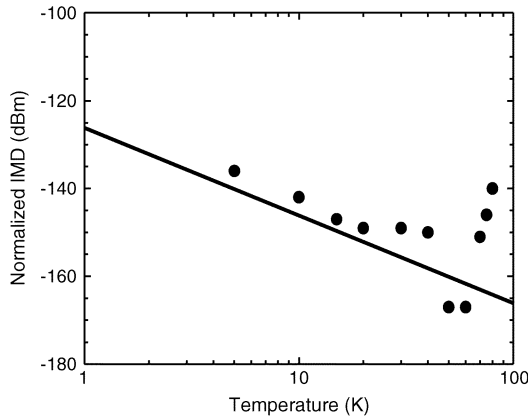


Fig. 6. IMD as a function of temperature for the 1200-nm-thick single-layer film. The line shows the  $1/T^2$  divergence that is characteristic of the intrinsic IMD in d-wave superconductors.

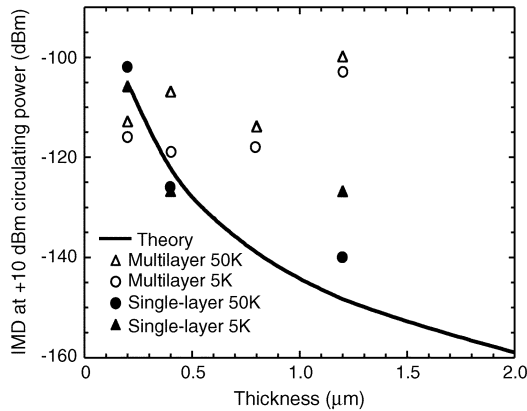


Fig. 7. Theory and experimental data for IMD. Solid line: the calculated IMD using the theory of the nonlinear Meissner effect [5]. Solid points are results for the single layer films and open points are for one set of multilayer films at 10 dBm circulating power, both at 5 and 50 K as indicated.

[8]. The exhibition of intrinsic nonlinearity indicates that the film is of high quality. Films with large numbers of structural defects do not show the intrinsically caused low-temperature increase [8], [9].

It is to be expected that at the same values of circulating power a thicker film has lower IMD because the current density is lower as the film grows thicker. However, the nonuniform current density and nonlocal electrodynamics must be considered. The microwave current is confined to the surface with a decay constant of the London penetration depth. In addition it has been found that nonlocal electrodynamics must be applied to the calculation of the nonlinear effects including the IMD [5]. While the confinement of the current to the surface would seem to lead to a constant current density independent of thickness, the nonlocal electrodynamics leads to stronger than expected dependence on thickness. The nonlocality implies that the average current density is the controlling parameter for the nonlinear effects. Thus

$$P_{IMD} \sim \left( \int dA \frac{I^2}{t^2} j^2(x, y) \right)^2, \quad (5)$$

where the integral is over the cross-sectional area of the strip,  $I$  is the total current,  $t$  is the thickness and  $j(x, y)$  is the current density; this expression is derived in detail in [5].

Fig. 7 shows the comparison of the calculated and measured dependence of IMD on width. Plotted is the IMD at 10-dBm circulating power for films of varying thickness at 5 and 50 K for single layer films and multilayer films where the YBCO layer thickness is 100 nm. The single-layer films show a decrease in IMD that is consistent with the calculation except that at a thickness greater than  $1 \mu\text{m}$  the decrease is smaller than predicted by the theory. The deviation from the theory is most likely due to a decrease in film quality as the films get thicker. The multilayer films show little or no decrease in IMD as the films get thicker. This is probably due to an even stronger decrease in film quality as the films get thicker and the number of multilayers increases, as shown in Fig. 3.

#### IV. CONCLUSION

We have fabricated high-quality YBCO films with various thicknesses by PLD using multiple targets. Thickness-dependence of crystalline quality and IMD for single-layer and multilayer films provided evidence that there is a strong correlation among between crystalline quality and IMD for YBCO films. The calculated IMD data using the nonlocal theory fitted well with the measured IMD data for single-layer films, indicating that achieving high-crystalline quality without degradation is critical for YBCO films of thickness greater than  $1 \mu\text{m}$  to show an intrinsic nonlinear behavior at microwave frequencies.

#### REFERENCES

- [1] D. E. Oates, S. H. Park, and G. Koren, "Observation of the nonlinear Meissner effect in YBCO thin films: Evidence for a d-wave order parameter in the bulk of the cuprates," *Phys. Rev. Lett.*, vol. 93, p. 197001, 2004.
- [2] D. E. Oates, S. H. Park, D. Agassi, and G. Koren, "Temperature dependence of intermodulation distortion in YBCO," *Supercond. Sci. Tech.*, vol. 17, pp. S290–S294, 2004.
- [3] D. Agassi and D. E. Oates, "Nonlinear Meissner effect in a high temperature superconductor," *Phys. Rev. B*, vol. 72, p. 014538, 2005.
- [4] D. Agassi and D. E. Oates, "Nonlinear Meissner effect in a d-wave superconductor: Extension to all orders of the vector potential," *Phys. Rev. B*, vol. 74, p. 024517, 2006.
- [5] D. E. Oates, D. Agassi, E. Wong, A. Leese de Escobar, and K. Irgmaier, "The nonlinear Meissner effect in a high-temperature superconductor: Local vs. nonlocal electrodynamics," *Phys. Rev. B*, vol. 77, p. 214521, 2008.
- [6] Z. Chen, F. Kametani, S. I. Kim, D. C. Larbalestier, H. W. Jang, K. J. Choi, and C. B. Eom, "Influence of growth temperature on the vortex pinning properties of pulsed laser deposited  $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$  thin films," *J. Appl. Phys.*, vol. 103, p. 043913, 2008.
- [7] K. J. Choi, H. W. Jang, and C. B. Eom, in preparation.
- [8] D. E. Oates, "Overview of nonlinearity in HTS: What we have learned and prospects for improvement," *J. Supercond. Novel Magn.*, vol. 20, pp. 3–12, 2007.
- [9] D. E. Oates, S. H. Park, D. Agassi, G. Koren, and K. Irgmaier, "Temperature dependence of intermodulation distortion in YBCO: Understanding nonlinearity," *IEEE Trans. Appl. Supercond.*, vol. 15, pp. 3589–3596, 2005.
- [10] D. M. Sheen *et al.*, "Current distribution, resistance, and inductance for superconducting strip transmission lines," *IEEE Trans. Appl. Supercond.*, vol. 1, pp. 108–115, 1991.
- [11] H. W. Jang *et al.*, Mechanism for the In-Plane Texture Improvement of  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  Films on Biaxially Textured MgO Templates for Coated Conductors to be published.
- [12] H. Xin *et al.*, "Third-order intermodulation distortion in YBCO grain boundaries," *Phys. Rev. B*, vol. 65, p. 214533, 2002.