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L. Chow
University of Central Florida

J. Chen
University of Central Florida

V. Desai
University of Central Florida

K. Sundaram
University of Central Florida

S. Arora
University of Central Florida

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Fractal formation of a Y-Ba-Cu-O thin film on $SrTiO_3$

L. Chow and J. Chen*
Department of Physics, University of Central Florida, Orlando, Florida 32816

V. Desai
Department of Mechanical Engineering, University of Central Florida, Orlando, Florida 32816

K. Sundaram
Department of Electrical Engineering, University of Central Florida, Orlando, Florida 32816

S. Arora
Department of Mechanical Engineering, University of Central Florida, Orlando, Florida 32816
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Fractal formation has been observed after thermal annealing of the rf-sputtered Y-Ba-Cu-O thin film on $SrTiO_3$ substrate. Through energy-dispersive x-ray analysis, it was found that the composition of the fractal was $YBa_2Cu_3O_x$ and the surrounding film composition was $Y_2Ba_2Cu_3O_x$. The fractal dimensions D ranging from 1.26 to 1.65 were obtained using the standard sandbox method with different thresholds.

Fractal formation has been an active field of research in recent years.[1-3] Various computer models[4-6] have been investigated and many experimental systems[7-12] like ultrafine smoke particles, porous sandstones, and electrolyte metal deposits have been studied. In this paper, we report the first observation of fractal growth of the high-temperature superconducting Y-Ba-Cu-O thin film on $SrTiO_3$ substrate.

The Y-Ba-Cu-O thin films were deposited by rf sputtering from a 2-in.-diam single composite 1:2:3 target onto a heated $SrTiO_3$ substrate. The target-substrate distance was kept at 2.5 cm, and the rf input power was usually set at 100 W. The typical thickness of the film was of the order of 0.5 to 1 μm. During deposition the sputtering chamber was kept at 20–30-mtorr pressure for an Ar-O_2 mixture of 2:1 ratio. The substrate was heated at 300 °C and was monitored by a type-K thermocouple. Due to the negative ion bombardment, the composition of the film was not homogeneous across the substrate. This resputtering phenomena in the Y-Ba-Cu-O single target has been reported before.[13] Details of our results on the composition variation will be reported elsewhere.[14] The as-deposited films were brownish in color and showed no features under the optical microscope, except the gradual variation from dark brown to light brown color. The resistance of the as-deposited film showed semi-insulating behavior. After thermal annealing in air at 870 °C for a half hour, fractal formation of Y-Ba-Cu-O was clearly observed under the optical microscope.

In Fig. 1, optical micrographs of different regions of the substrate are shown which clearly indicate the different growth stages of the fractals. In Fig. 1(a) the initial nucleation of the Y-Ba-Cu-O is clearly seen. The size of the nucleation seed is about 1 μm. In Fig. 1(b), the treelike growth of the fractal is shown, it shows the striking similarity to the computer simulation done by Voss.[15] This anisotropic growth of the fractal reflects the composition variation of our deposited films. In Fig. 1(c), a typical fractal is shown. Again this fractal still showed slight anisotropic shape. The size of these typical fractals is on the order of 100 μm. Based on our experience that the annealed 1:2:3 thin film is black in color, we suspect that the dark fractal region is filled with the superconducting Y-Ba-Cu-O granular network embedded in the nonsuperconducting oxide. Through x-ray energy dispersive analysis, it is confirmed that the composition inside the fractal patterns is close to 1:2:3 and the surrounding film composition is close to 2:2:3. The different growth stages showed in the same substrate here is an indication of the strong composition variation across our substrate. We did an x-ray analysis of a Y-Ba-Cu-O film deposited on a stainless-steel substrate. The composition variation across the substrate is shown in Fig. 2. This variation of the composition is due to the selective negative ion resputtering mentioned above. We can see that while at a radius of larger than 3 cm the composition is roughly constant, the Y and Ba concentrations showed dramatic change between the 2- and 3-cm radius. Our $SrTiO_3$ substrate was placed at this distance during deposition, which explains the strong composition variation of the as-deposited films.

The fractal growth of the 1:2:3 phase thin film on $SrTiO_2$ was also studied through the scanning electron microscopy (SEM). In Fig. 3(a), many square-shaped single-crystal granulars with the size of 1 μm or more are closely connected to form the fractal networks. In view of their morphology and the color, such granulars are believed to be $YBa_2Cu_3O_{7-x}$ (1:2:3) superconducting crystallites with common c axis, but different a or b axis. It is likely that such crystallites grew epitaxially from the $SrTiO_3$ substrate because the substrate is (001), whose lattices are well matched with the 1:2:3 phase. The area between the fractal networks is filled with many smaller amorphous granulars, which may be in the glass state or other nonsuperconducting phases.

(a) (b) (c)

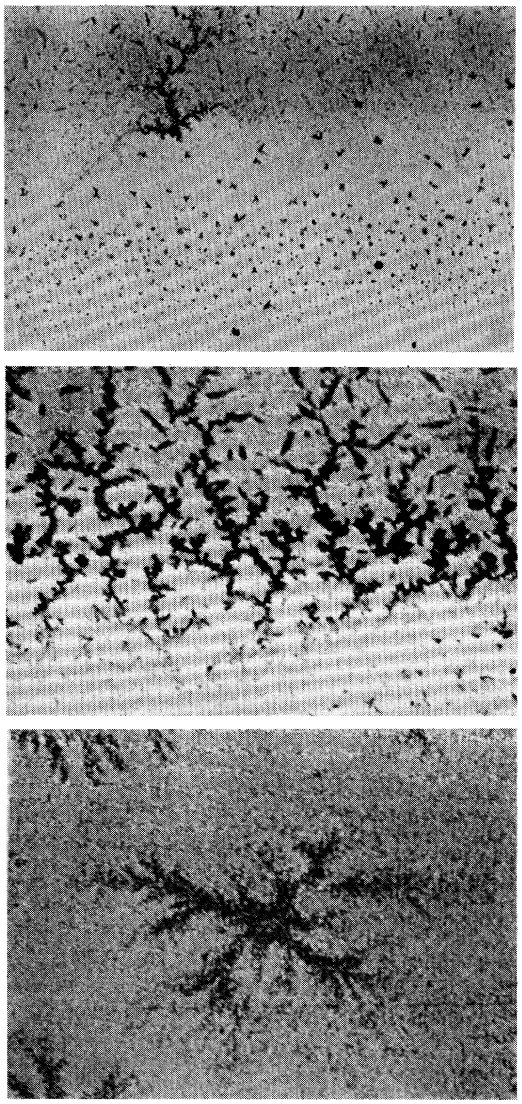

FIG. 1. Optical micrographs of different stages of the fractal growth: (a) nucleation, (b) treelike structure, and (c) a typical fractal.

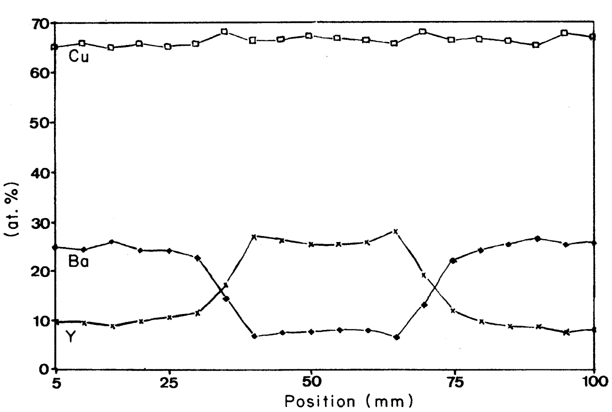

FIG. 2. Composition variation of the as-deposited Y-Ba-Cu-O thin film on stainless steel. The target composition is 1:2:3, and the center of the target is around 50 mm.

(a) (b)

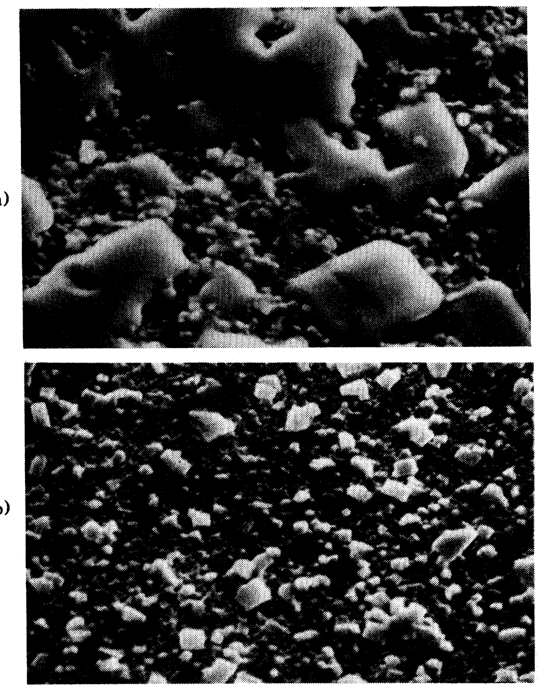

FIG. 3. SEM micrograph of the Y-Ba-Cu-O film on $SrTiO_3$. (a) Inside the fractal pattern, the epitaxially grown 1:2:3 phase networks are clearly seen; (b) outside the fractal pattern, it mainly consists of unconnected small crystallites.

Figure 3(b) showed the film outside the fractal patterns which is composed of many unconnected small crystallites. There are at least two different kinds of crystallites. One is the 1:2:3 phase single crystallite, and the other may be the Y_2BaCuO_x (2:1:1) phase. It was reported[16] recently that the 2:1:1 phase is also easily nucleated and grew at the temperature of 900 °C.

The SEM observations agreed with our x-ray analysis given above. Namely, the fractals are made of many small and well-connected 1:2:3 phase crystallites.

The micrograph of the fractal pattern of the Y-Ba-Cu-O film was digitized with a camera into a 512×512 matrix. The intensity of each pixel ranges from 0 (black) to 90 (grey background). Using a standard threshold detection method,[10] we assign the number 1 to sites with intensity less the threshold, and the number 0 to sites with intensity larger than the threshold. The 0-1 two-dimensional array generated with a threshold equal to 40 is shown in Fig. 4, which corresponds to the micrograph shown in Fig. 1(c). Since the fractal dimension can be defined as

$$M(L)=L^D, \qquad (1)$$

we use the simple sandbox method[17] to calculate the fractal dimension D. We found that the fractal dimension depends very sensitively on the threshold we chose. In Fig. 5, the log-log plots of $M(L)$ vs L for thresholds = 40, 50, and 60 were shown. For $L < 15$, a slope of 2 is obtained for all three thresholds; this corresponds to the completely filled center black region of the fractal. For $L > 15$, we

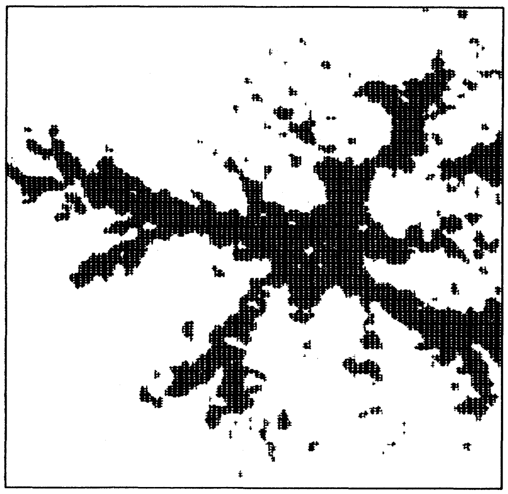

FIG. 4. A digitized two-state map of the fractal generated with a threshold equal to 40. This corresponds to the fractal shown in Fig. 1(c).

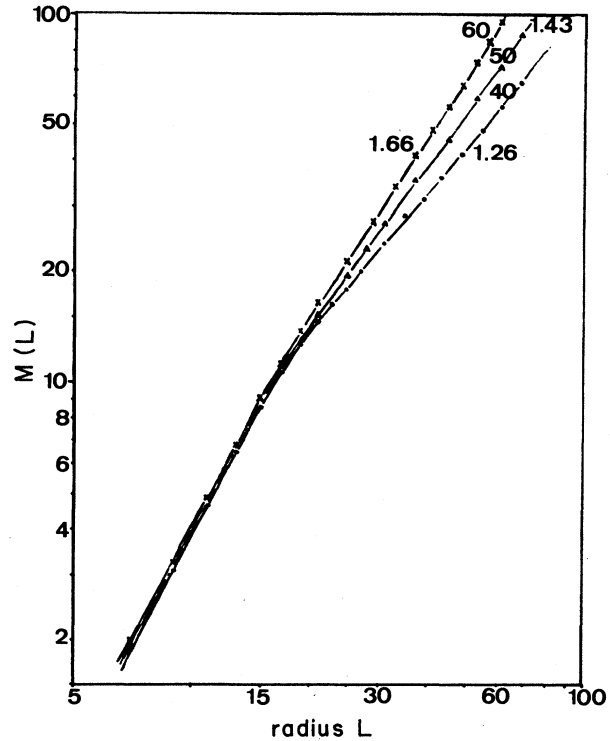

FIG. 5. Log-log plot of mass $M(L)$ vs radius L for the fractal shown in Fig. 1(c). Slopes equal to 1.26, 1.43, and 1.66 for thresholds equal to 40, 50, and 60.

obtained $D = 1.26$, 1.43, and 1.65 for thresholds = 40, 50, and 60. A detailed description of the fractal dimension analysis of the different fractal growth stages will be given elsewhere.[18]

There are various growth models describing the fractal aggregation, which give different values for the fractal dimension. Here we will discuss two basic models which may be related to our work. In the diffusion-limited aggregation model, the cluster is formed through random diffusion of particles. A sticking coefficient of one is assigned, such that it is an irreversible process. In the reaction-limited aggregation, the formation of the cluster is governed by chemical reactions. In our system, it is complicated by the composition variation as mentioned above, and the fact that it is a multicomponent system; we feel that both mechanisms may be important in the formation of clusters. Namely, at the early stage, there are local regions where compositions are close to 1:2:3, and due to the fact that the substrate has a good lattice match with the 1:2:3 phase, epitaxial growth of the YBa_2Cu_3O 1:2:3 phase is likely to happen. Once nucleation is formed, the further growth of the cluster depends on the neighboring composition and interdiffusion of the various components. Since the 1:2:3 phase is a more stable phase in the as-deposit mixed oxide film, the latent heat released, as the cluster grows, will further aid the growth. Presently, we feel that further experimentation is needed to elucidate the details of the physical mechanism of the growth.

Finally, we want to mention that in a recent Letter,[19] the authors relate the superconductivity to the fractal dimension of the bulk Y-Ba-Cu-O sample. They measured the fractal dimensions of the superconducting grains, nonsuperconducting grains, and the cavity in bulk sample. They found that fractal dimensions range from 1.06 to 1.31, depending on the thermal treatment of the sample. It is not clear whether their measurement is related to our work at the present moment.

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*Permanent address: Department of Physics, Nanjing University, Nanjing, People's Republic of China.

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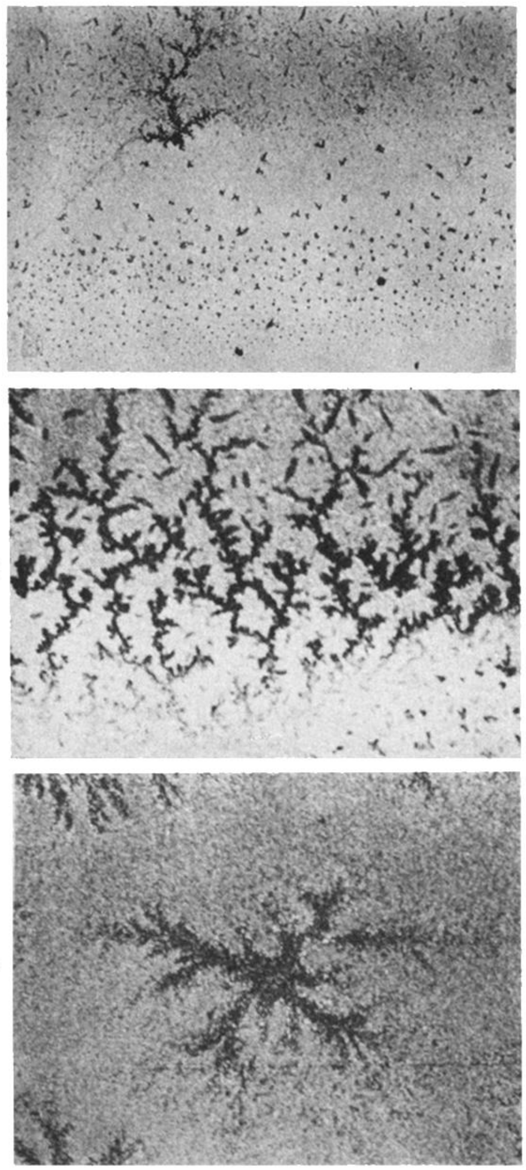
(a)

(b)

(c)



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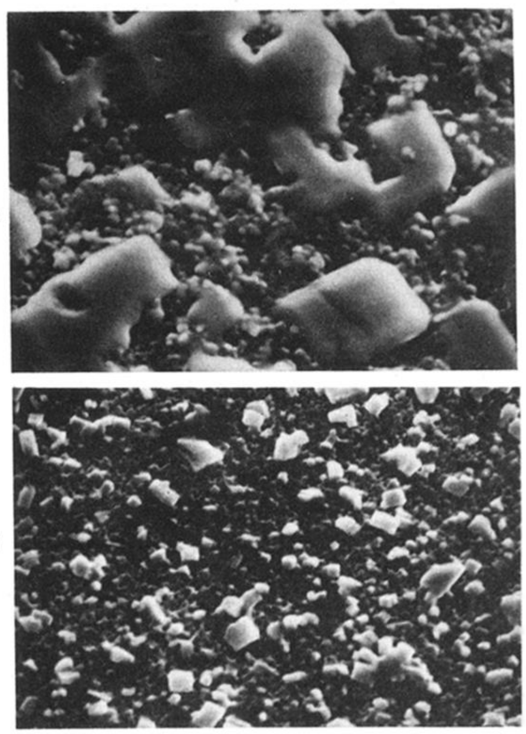

(a)

(b)

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