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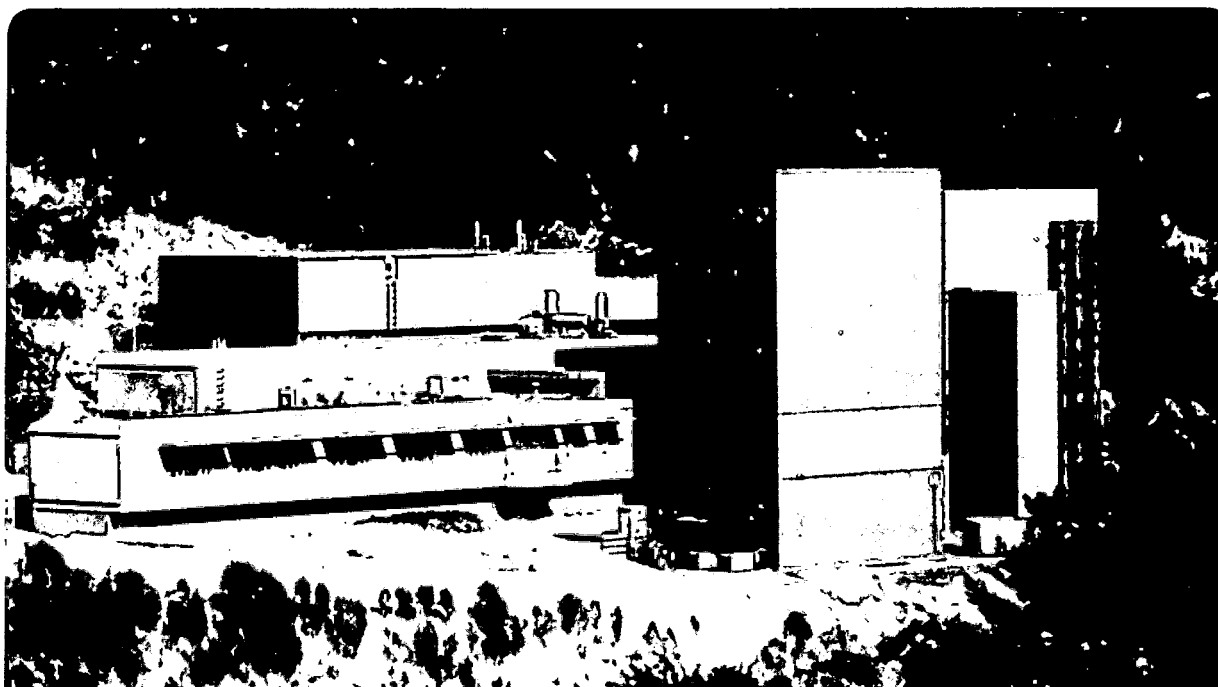
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### Computer Simulation of Growth Mechanisms in Y-Ba-Cu-O Thin Films

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**Computer Simulation of Growth Mechanisms  
in Y-Ba-Cu-O Thin Films**

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## ABSTRACT

The growth of epitaxial thin films of  $\text{YBa}_2\text{Cu}_3\text{O}_7$  is studied by means of computer simulations of the deposition and diffusion of Y, Ba, and Cu oxide particles. The evolution process is modeled by means of a three-step Monte Carlo technique incorporating one sorption deposition mode and two modes of film annealing addressing surface and bulk diffusion. A systematic study of the effects of deposition rate and substrate temperature during in-situ film fabrication reveals that the kinetics of film growth can readily dominate the structural formation of the thin film leading to dramatic morphological transitions as a function of deposition conditions alone.

**KEYWORDS:**  $\text{YBa}_2\text{Cu}_3\text{O}_7$  Superconductor Thin Film, Computer Simulation, Kinetics of Crystal Growth

## INTRODUCTION

Some of the most exciting applications of the  $\text{YBa}_2\text{Cu}_3\text{O}_7$  family of high  $T_c$  oxide superconductor are in the field of microelectronics; in particular in superconducting quantum interference devices (SQUID) and in integrated circuit interconnections[1]. These devices exploit the ability of the thin film process to produce highly oriented and textured material. However, the thin film deposition process is notoriously complex[2]. Some of the many possible variables include substrate type, orientation, and defect density, substrate or ambient temperature, deposition technique, source ratios, deposition atmosphere, deposition rate, and post annealing processes. While the large number of thin film studies are providing insight into the general effects of specific deposition conditions on  $\text{YBaCuO}$  thin film growth, it is difficult to design an ideal experiment that precisely controls all but one of the many variables to allow investigation of the effect of a particular parameter on film growth. Since thin film growth is a non-equilibrium process, the evolution of the system is expected to be strongly influenced by impurities and by surface inhomogeneities[3]. As a result, it is difficult to interpret experimental kinetic data as attributable to a specific macroscopic parameter since microscopic effects can easily dominate. In addition, most characterization techniques lack either sufficient time or spatial resolution necessary to allow study of the kinetic evolution of thin film structures. In many other systems, the problem of characterizing non-equilibrium evolution has been addressed by "enhancing" the resolution of experimental techniques with the effectively "infinite" time and spatial resolution of computer simulation. Simulation allows both the development and evaluation of microscopic theories of kinetic evolution and a degree of control over the "experimental" conditions inaccessible by experimental methods. Thus, for the purpose of providing an understanding of the critical kinetic factors dominant in the deposition of Y-Ba-Cu-O superconducting films, a technique for the computer modeling of thin film deposition and growth is introduced and its theoretical and experimental basis briefly described. The ability of this technique to model established trends is examined and results of the effects of deposition conditions on film characteristics are reported.

## THEORETICAL DEVELOPMENT

In order to mirror the rich range of fabrication conditions afforded by deposition techniques, the simulation must be based on the predominant microscopic mechanisms operating during thin film deposition. The current technique is inspired by the Kossel or "Solid-on-Solid" (SOS) model of crystal growth[4,5] in that incident particles are adsorbed normal to the substrate, precluding the formation of voids or overhangs, and only nearest neighbor pair interactions are employed. The simulation utilizes a three-part extension of the Monte Carlo technique to model deposition (sorption), surface diffusion, and bulk realignment of deposited species. As the Y-Ba-Cu-O system can be described by a ternary phase diagram of yttrium, barium, and copper oxides[6], the simulation "deposits" variable ratios of these oxides particles.

Within a unit cell of a  $\text{YBaCuO}$  phase, each atom can be assigned a type and an orientation with respect to the substrate which reflects the crystallographic orientation of the domain it participates in within the thin film. In addition to the three oxide types, two additional particle types are used to represent the substrate and "unoccupied" sites. These unoccupied particles correspond to sites above the current surface of the film and are used to account for orientation specific surface energies. Amongst the yttrium, barium, and copper oxide particles, the pair interactions are taken over the six nearest neighbor sites in the lattice: four "in-plane" (001) sites and two "out-of-plane" [001] sites. These interactions are dependent on both type and direction and their magnitudes are chosen to reflect the in-plane clustering and out-of-plane layer ordering experimentally established for the layered Y-Ba-Cu-O superconductors. Oxide-substrate interactions vary in the same manner allowing the modeling of different substrate types and orientations. Thus the Hamiltonian describing the configurational energy of microstates arising under the simulated deposition

process is composed of a generalized Ising model reflecting surface, vacancy/surface, and oxide interactions, coupled with a Heisenberg spin interaction reflecting the variation of interactions with particle misorientation. This Hamiltonian can be written in the compact form:

$$H = - \sum_{\langle i,j \rangle} V_n(n_i)n_jn_j - \mu_i \sum_{\langle i,j \rangle} n_i - \sum_{\langle i,j \rangle} V_s(\theta_{ij})(s_i \cdot s_j)$$

where  $n_i = 1, 2, \dots, 5$  denotes the type at site  $i$ , and  $|s_i| = 1$ . The chemical potential terms,  $\mu$ , in the Ising model component of the Hamiltonian, while taken to be zero in this study, can be used to describe different deposition atmospheres above the film with respect to the different metallic species in the film. In the current study, particles interact via symmetrical pair interactions  $V_n, V_s$  only. Thus a logical enhancement of this model might include multi-spin interactions.

Numerous attempts have been made to relate the bonding properties of Y-Ba-Cu-O to its underlying electronic structure. For example, the effective pair interactions ("exchange energies") in the CuO basal plane have been derived from first principles LMTO calculations[7]. Using phenomenological or electron-gas theories, other authors[8,9] have determined bond energies (pair and many body potentials) that stabilize the structure. These interactions have been mainly used in static lattice simulations or phonon calculations and were adjusted to fit certain experimental data. Even so, parameters derived by different groups may differ considerably and in some cases include unphysical interactions. To our knowledge, no molecular dynamics simulations of the type of non-equilibrium and off-stoichiometry phenomena inherent to film evolution have been reported. However, the large amount of experimental work available for this system can be used to derive interactions which stabilize the  $YBa_2Cu_3O_7$  structure under this model of thin film deposition. Doing this, one can then use these interactions to study the system at finite temperatures and under non-equilibrium conditions. In order to derive values for pair interactions for this system, a number of consistent characteristics of the Y-Ba-Cu-O system were utilized. These are the layered plane nature along the [010] type directions, and the interatomic cation ordering occurring along the [001] direction. This is a strong indication that interactions amongst species of the same type are attractive "in-plane" or in the [100] and [010] directions. In addition, the ordering of the layers in the [001] direction suggests an attractive interspecies interaction along this axis. The actual mechanism responsible for layer ordering in the [001] direction is most likely complex involving long range interactions of extent at least equal to that of the unit cell. However, in this first attempt at modeling thin film growth, a simple statistical approximation to this is employed. In the model described, a 123 unit cell is built via the deposition of a sequence of Cu, Ba, and Y oxide blocks (the  $CuO_2$  plane is incorporated into the Ba and Y particles in an unspecified manner). A correct ordering, as observed experimentally, would indicate attractive Cu-Ba and Ba-Y symmetrical pair interactions along the [001] direction, all others being repulsive. In addition, [001] direction self interaction energies (Cu-Cu, Ba-Ba, Y-Y) are modified by a chemical potential term,  $-\mu_i n_i$ , included to represent the influence of partial pressures of the  $i$ th component in the deposition atmosphere. The chemical potentials are normalized such that a stoichiometric 1:2:3 composition corresponds to  $\mu_i (i \in \{Cu, Ba, Y\}) \equiv 0.0$ . In a lattice gas representation of pair interactions, attractive interactions are taken to be negative in sign and repulsive interactions positive. This representation leads to interactions between five particles (Y, Ba, Cu, Substrate, and Vacant types) in the three  $\langle 100 \rangle$  directions. However, the potential 75 interaction tensor reduces by symmetry and the ground state analysis for the current simulation to three distinct magnitudes of interaction in the following fashion. Pair interactions are taken to be symmetric, via the definition of effective pair interactions, reducing the set to 25, and the action in the [100] and [010] are taken to be identical during deposition (an assumption that invokes the observation that oxygen is not ordered during the deposition leading to an effectively tetragonal material). In addition, the canonical assumption is made that interactions in equivalent crystallographic directions, whether attractive or repulsive, have the same magnitude.

The simulation model employs three excitation mechanisms which model deposition, surface redistribution, and bulk realignment of oxide particles. The deposition step, an inherently open process, is carried out in the grand canonical scheme with Kawasaki dynamics[3]. Sites on the surface of the existing film are randomly selected and a random type is assigned to the oxide weighted by the selected cation deposition ratio for the current simulation run. Deposited particles have an initially undetermined crystallographic orientation, but the action of the Heisenberg component of the Hamiltonian favors epitaxial growth. The configurational energy of the attempted deposition is calculated from the above Hamiltonian and deposition is accepted via a classical Monte Carlo weighting based on the grand canonical density function. The rejection of a deposition attempt corresponds to the phenomenon of beam reflection. The surface redistribution step, involving the exchange of nearest neighbor surface particles and vacancies, conserves the total number of particles in the film and is thus carried out in the canonical ensemble with Glauber dynamics[3]. An adjacent surface particle-vacancy pair is selected, and the change in configurational energy for an exchange of position is calculated. Exchanges are accepted with a probability based on the canonical density function. Finally, the bulk realignment step involves the attempted orientation change of an oxide particle within the film. As the "orientation" property is not conserved under this step, orientation changes are calculated in the grand canonical scheme with Kawasaki dynamics. Trial orientation realignments are accepted with a probability weighted by the grand canonical density function.

The deposition rate for the simulation is defined as the number of deposition steps attempted divided by the total number of simulation steps attempted (deposition, surface redistribution, and bulk realignment steps). Thus, a deposition rate equal to 1 corresponds to pure deposition and no "annealing." In this way, in-situ vs. ex-situ thin film deposition schemes are modeled. During in-situ deposition, deposition and "annealing" (surface redistribution and bulk realignment) are carried out simultaneously in a ratio corresponding to the deposition rate selected for the current simulation. By contrast, during ex-situ deposition, all the deposition steps are carried out first, often producing

amorphous films, after which the annealing steps are performed.

The resulting simulation technique allows the simulation of a wide range of deposition conditions. In this study, the substrate is chosen to be atomically smooth, defect free, and its interaction with the deposited material constant, attractive, and independent of crystallographic orientation so as to effectively eliminate the influence of the substrate from the study of the effects of other parameters. Deposited material is also taken to be fully oxygenated. In addition, the orientation vector of the deposited particles is constrained to the  $\langle 100 \rangle$  unit cell directions. Studies are in progress which remove these constraints in the study of substrate variations and the development of the full range of grain boundary and surface orientations.

## SIMULATION RESULTS

Many hundreds of simulated films were produced under various deposition conditions. All films simulated employed lattice sizes containing on the order of  $1 \times 10^6$   $\text{YBa}_2\text{Cu}_3\text{O}_7$  unit cells. For a simulation lattice with toroidal boundary conditions in the [100] and [010] directions created such that its lateral dimensions are an order of magnitude greater than its vertical extent, this produced films approximately 25 nm thick. All films reported here were deposited with a cation ratio of 1:2:3 (Y:Ba:Cu) with a slight shortage of barium in order to produce stoichiometric  $\text{YBa}_2\text{Cu}_3\text{O}_7$  thin films. The discovery that a Barium deficient deposition stoichiometry leads to more homogeneous and defect-free films was made by literally "experimenting" with different stoichiometries. It is significant to note that an experimental study by Matijasevic et al.[10] confirms that a Ba-deficiency leads to thin films of higher quality. This strongly suggests that the granularity of deposition in this system is indeed on the level of more complex cation-oxide structures, as is assumed in this study. In such a scheme, each unit cell requires a Ba-Y-Ba ordering to successfully grow. In this sequence, a deficiency of Barium, the majority "unit", would control the deposition of Yttrium and lead to higher quality films.

As the orientation of particles is constrained to the cube directions, individual domains within the film are found to fall into two general classes: domains whose unit cell [001] 'c' axis was oriented perpendicular to the substrate, or 'c' type, and domains whose 'a' or 'b' axis was oriented perpendicular to the substrate, or 'a' type. As the  $\text{YBa}_2\text{Cu}_3\text{O}_7$  material is assumed to have the tetragonal structure during growth, no distinction is made between 'a' and 'b' type growth.

## SURFACE MORPHOLOGY

By varying the conditions under which the films were produced, a broad range of orientation types and surface morphologies were observed. Ex-situ films of the 'a' type show surface morphologies composed of a network of [100] and [010] oriented lenticular shaped interlocking "bars" of Y-Ba-Cu-O material. The surface of such a film is shown in Figure 1a. The size of such domains was seen to coarsen upon increasing annealing times. Deep "grooves" in the film surface sometimes are found to occur within single domain regions indicating an anti-phase relation between regions separated by this boundary. This type of defect arises as the anti-phase boundary propagates upward during film growth and indicates a region of unfavorable bonding condition for all metallic species. This anti-phase surface feature is expected to be less pronounced in real films as it is on the order of a unit cell width and should be obscured by surface relaxation which is not explicitly handled by static lattice simulation. Overall, ex-situ 'a' type film surfaces were found to be very "rough" and characterized by rectangular shaped protrusions of 'a' type material in which the height of the film surface from the substrate varies markedly from point to point. In contrast, ex-situ films of the 'c' type (see Figure 1b) displayed a much more consistent surface height. The overall shape of the 'c' type domains was more circular in nature than the interlocked 'a' type domains, producing a "pebbly" morphology.

Domains of anti-phase relation were similarly separated by deep grooves in the film surface. Surfaces of in-situ grown films displayed a much less dramatic surface structure. Surfaces were generally atomically smooth (except under high deposition rates or low temperatures) but domain and anti-phase boundaries were decorated with surface defects and depressions. These decoration patterns were often sufficient to identify the surface type as the pattern inscribed would be orthogonal and rectangular in nature for 'a' type surfaces and more curvilinear in nature for 'c' type surfaces.



Figure 1: Surface structures produced during ex-situ thin film deposition simulation. (a). 'a' type film displaying characteristic interlocking orthogonal domains.

(b). 'c' type surface displaying characteristic curvilinear domains and less dramatic relief than that of the 'a' type film.

Numerous thin film studies describe surfaces or include surface micrographs of ex-situ[11-14] or in-situ[15,16] deposited films which display completely analogous features as those produced by the simulation.

Under specific conditions, adjacent domains of 'c' type and 'a' type material can be observed to grow. The growth rate anisotropy between the 'a' and 'c' directions of the Y-Ba-Cu-O unit cell is apparent in the surface characteristics of such films. The greater growth rate of the 'a' type domains leads to regions of the film surface that are notably greater in vertical extent than adjacent 'c' type domains. The anisotropy of growth rates between 'a' and 'c' type surfaces can be understood by an analysis of the surface dependence of the adsorption rate,  $r_a$ . The adsorption rate can be written in the following form:

$$r_a = S^\alpha(\phi)F$$

where  $S^\alpha(\phi)$  is the sticking coefficient as a function of surface coverage,  $\phi$ , and  $F$  is the incident flux rate[17]. The superscript,  $\alpha$ , is taken to be 1 for this study to represent non-dissociative adsorption. The sticking coefficient, in turn depends on coverage and the activation energy for adsorption,  $E_a$ , by:

$$S^\alpha(\phi) = S^\alpha(0)(1 - \phi)^\alpha = S_0^\alpha \exp(-E_a/k_B T)$$

where  $S^\alpha(0)$  is the sticking coefficient at zero coverage, and  $S_0^\alpha$  is the pre-exponential factor[17]. The activation energy for adsorption is composed of two terms; structural coordination and, as this is a multi-component deposition process, chemical coordination. An atomically rough or stepped surface will provide a higher density of kinks and ledges where an adsorbate will find a maximum number of bonds to join during adsorption, increasing the activation energy for the process. Since this simulation is constrained to the cube faces, the structural contribution is expected to be equal on both the 'a' and 'c' type surfaces. However, the average chemical coordination density on a surface will vary drastically for an 'a' or 'c' type surface. For a random but stoichiometric flux of metallic deposition species, an 'a' type surface provides favorable sites for deposition of all species within a unit cell distance whereas a 'c' type surface provides at any given time favorable deposition sites for only a subset of the stoichiometric flux. Thus particles captured by a 'c' type surface are expected to have longer residence times, dependent on the density of ledges, before finding a high activation energy adsorption site than a particle captured by an 'a' type surface. The anisotropy in growth rates is thus expected to scale as the number of atomically distinct layers in the 'c' direction of the unit cell of a given phase. Thus the sticking coefficient, through contributions via chemical coordination, and, hence, the growth rate is expected to be higher on 'a' type surfaces than on 'c' type surfaces in  $YBa_2Cu_3O_7$ .

#### MORPHOLOGICAL TRENDS

To investigate the effects of deposition rate and substrate temperature on the growth and orientation of in-situ grown films, a systematic study was performed under conditions where 'c' type growth is energetically preferred. In these studies, the effects of the substrate and the deposition atmosphere were held constant. Figure 2 presents four cross-sections of films produced at constant substrate temperature of  $k_B T/V_B = 0.8$  (temperature units normalized by the in-plane interaction energy between  $BaO$  particles,  $V_B$ ) for various deposition rates. At a low deposition rate equal to 0.0001, the defect free, 'c' type film of Figure 2a is produced. At a slightly higher deposition rate of 0.001, the film produced is a defect free 'a' type film (Figure 2b) indicating an apparent transition from 'c' to 'a' type film texture. At higher deposition rates, a mixed 'c' and 'a' type film is produced which includes an assortment of defects (Figure 2c). These include point defects, antiphase boundaries, columnar grains, and near (110) type boundary between the 'a' and 'c' type domains. Finally, at a very high deposition rate (0.25), a highly defected film with many small domains of orthogonal and antiphase relation is produced (Figure 2d). In order to further investigate the apparent 'c' to 'a' type transition as a function of deposition rate, a more systematic study was performed to quantify the variation of the volume fraction of 'c' type film produced at various deposition rates. The results of this study are shown in Figure 3. The points corresponding to the snapshots of Figure 2 are labeled in Figure 3 as "a"- "d". At high deposition rates, the distribution of sites in 'c' and 'a' type orientations is nearly equal, reflecting the nearly amorphous nature of films produced under these conditions. As the deposition rate is decreased, the fraction of 'c' type growth gradually increases. In this range, the growth of films appears to be nucleation limited and films consist primarily of many small domains of 'c' type material. Note that the surface mobilities, also plotted in Figure 3, remain essentially equal throughout this region reflecting that a nucleation and not growth mechanism is predominant. However, as the deposition rate is decreased below 0.01, a sudden transformation to 'a' type growth occurs. This transformation occurs with a corresponding drop in the surface mobility of 'a' type domains, and an increase in the mobilities of 'c' type domains. This bifurcation of the surface mobilities heralds a transition to growth limited kinetics and the lower 'a' surface mobilities reflect that more particles are adsorbing to 'a' type surfaces. While equilibrium considerations indicate that 'c' type growth should still prevail, the adsorption rate anisotropy between the 'a' and 'c' faces of the  $YBa_2Cu_3O_7$  material provides 'a' type growth with a kinetically more favorable route. It is felt that most thin film experiments are carried out in this regime. At lower deposition rates, the trend is again seen to reverse. At this second transition, substrate effects now lead to the nucleation of 'c' type domains, but the growth limited kinetics quickly lead to the formation of 'a' type growth thus producing the type of 'c' to 'a' type transformation through thickness in thin films which is commonly reported[11,12,15,16,18,19] and also observed in the simulation. As the deposition rate is decreased further, the growth proceeds via a two-dimensional layer mechanism under which the equilibrium free energy of formation of the 'c' type phase is great enough to overcome the kinetic path afforded to 'a' type domains by the growth rate anisotropy and only 'c' type grains nucleate and grow. Note that the 'a' and 'c' surface mobilities approach a constant value. A similar trend is found with the variation of the film orientation with temperature at a constant deposition rate of 0.001. High temperatures are again nucleation limited, and polycrystalline films of 'c' type domains prevail. As the temperature decreases, the growth anisotropy intercedes and 'a' type films become dominant with a corresponding crossover in surface mobilities. At very low temperature, the growth anisotropy is overcome by



the equilibrium free energy of formation for 'c' type material, and 'c' type grains prevail. Preliminary results for conditions under which 'a' type growth is energetically preferred do not show the 'a' to 'c' to 'a' transformation, as here, 'a' type is both the kinetically and energetically preferred phase.

The simulation detects two analogous kinetic morphological trends: transitions from 'c' to 'a' and back to 'c' type thin film growth upon either decreasing deposition rate (degree of supersaturation) or substrate temperature. It is worth reiterating that in the current study, the effect of the substrate is removed. Some experimental studies have suggested that the growth orientation of the film is dependent on the type of substrate material used. While the substrate is certainly expected to have a role in determining film growth, this simulation establishes that a variation of deposition rate or substrate temperature *alone* is sufficient to lead to differing growth morphologies. Various experimental studies provide evidence confirming or consistent with the results of this simulation. Matijasevic et al.[10], through a study of the intensity ratios of (002) to (006) x-ray diffraction peaks ( (002) indicates the presence of 'a'-axis material) for varying substrate temperature, show that the main orientation transforms from 'a' type to 'c' type at approximately 600°C. In addition, 'a' type material was detected from the transition at 600°C down to 530°C while 'c' type material was produced up to 750°C. Other studies have reported the effects of deposition rate on film morphology. In experiments performed at deposition rates below those in which 'a' type films had been achieved, Westerheim et al. repeatedly obtained 'c' type films[20].

The films used in the morphological studies were very thin and on the order of 4 nm thick. Thus the resulting films tend to be completely homogeneous through thickness which results in the sharp transitions from 'a' to 'c' type films. It is expected that these sharp transitions found in the variation with substrate temperature will "round" out for greater film thicknesses, reflecting substrate retained material of a subordinate orientation type in a given deposition range. In many ways, Figure 3 and its analogous plot for substrate temperature, could then be thought of as isothermal and isochronal sections taken through a classical TTT (Time-Temperature-Transformation) where time is replaced by deposition rate and the transformation altered to indicate the degree of 'a' or 'c' type film.

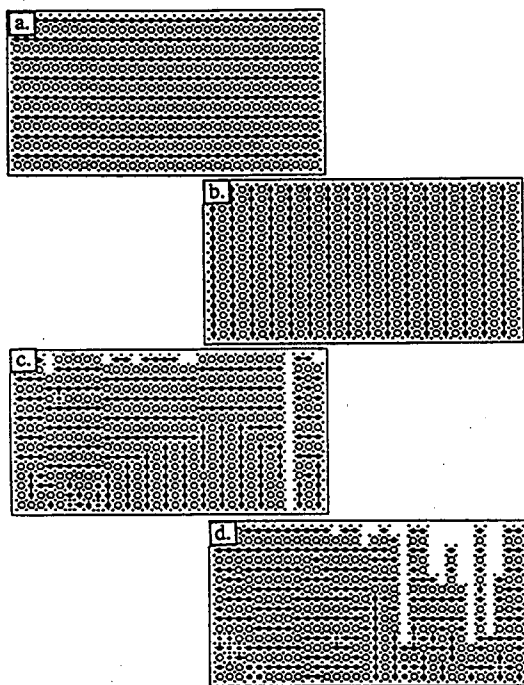


Figure 2: Cross-sections of in-situ films produced at constant simulation temperature  $k_B T/V_B = 0.8$  for different deposition rates. Substrate is not depicted but lies at the base of each cross-section. Open circles denote barium atoms, filled circles represent both copper and yttrium atoms (for graphical simplicity) and lines, when present, lie in the [100] and [010] directions to denote 'a' type film texture. (a). Defect free 'c' type film produced at deposition rate (DR) of 0.0001. (b). Defect free 'a' type film produced at DR = 0.001. (c). Defected film containing both 'a' and 'c' type variants displaying anti-phase boundaries, point defects, columnar growth, and grain boundaries produced at DR=0.01. (d). Highly defected film of primarily 'c' type displaying many small domains (DR=0.25).

#### ROLE OF SPIRAL DISLOCATIONS

While spiral growth, which originates through the elastic effects of a screw dislocation, can not be modeled in a pure

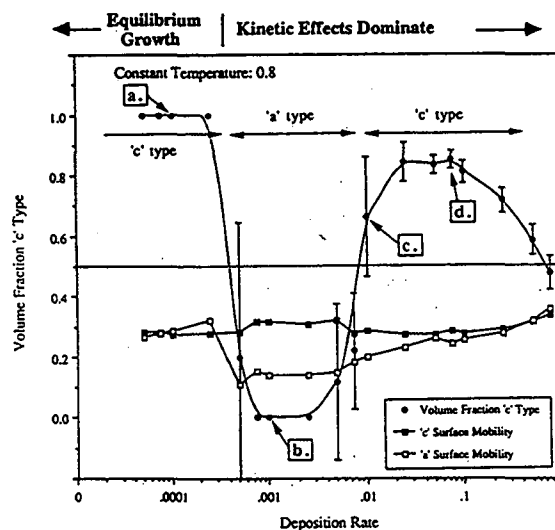


Figure 3: Plot of volume fraction of 'c' type film vs. deposition rate at constant substrate temperature of  $k_B T/V_B = 0.8$  displaying transitions from 'c' to 'a' type growth. Filled circles represent volume fraction of 'c' type material, filled squares represent surface mobility of particles on a 'c' surface, open squares, 'a' surface mobility. Each point represents the average of five simulated film depositions and error bars represent the standard deviation in the data.

static lattice simulation, a simulation of growth kinetics in the absence of elastic effects, coupled with classic phenomenological descriptions of spiral growth, can do much to elucidate how and under what conditions spiral growth occurs. It is well understood that spiral growth occurs under conditions of low surface saturation where few adatoms are available to act as nucleation sites for standard island, layer, or island/layer epitaxial growth modes, as has been noted in experimental studies in this system[21]. In this simulation, as in experimental studies, the degree of surface saturation corresponds directly to deposition rate. The observed morphological dependence on temperature and deposition rate can be phenomenologically understood in terms of classical growth mechanisms. Supersaturation leads to a preponderance of kinks and ledges which favors simultaneous island/layer growth. Low deposition rates lead to a pure layer growth and a 'c' type morphology in the absence of misfit dislocations or other screw dislocation nucleation sites. However, if misfit dislocations, islands, or surface steps were present, one can easily envision a situation where growth concentrates to the site of an existing screw dislocation leading to spiral growth. Grains originating via spiral growth would rapidly over-grow island/layer growth mechanisms under low saturation (low deposition rate) conditions. The effect of the addition spiral growth on the morphological observations in this study would be minimal as 'c' type growth is already the preferred morphology found at low surface saturations.

## CONCLUSION

A technique for the computer simulation of the thin film deposition and growth of YBaCuO is described. Typical surface morphologies for in-situ and ex-situ fabricated thin films are examined and found to be analogous to those commonly reported elsewhere. Films produced under certain conditions exhibit the commonly observed 'c' type to 'a' type orientation transformation through thickness. A systematic study of the growth of in-situ films under conditions where 'c' type film growth is energetically preferred is performed. It is found that kinetic factors can lead to broad ranges in the values of deposition rate and substrate temperature which give rise to 'a' type film. Such factors are asserted to be the mechanism producing commonly observed 'c' to 'a' type film crystallographic orientations through the film thickness. Finally, it is worth mentioning that although the present discussion was cast in terms appropriate for the Y-Ba-Cu-O system, the conclusions are expected to be generally valid for other superconducting thin films and other multicomponent materials systems.

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