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Fanlike aggregations on seeds by parallel ballistic flux: Experimental results and Monte Carlo simulations of the growth of three-dimensional Si structures

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Previously, it was shown by theories and computer simulations that intriguing fanlike aggregations on a seed could be created from a parallel ballistic flux under a highly nonequilibrium condition. In this paper, we report the creation of such fanlike structures on small size (∼150 nm in diameter) cylindrical seeds using a thermal evaporation technique. Monte Carlo simulations, based on a ballistic aggregation model with surface diffusion, were employed to quantitatively describe the characteristics of the fans, including the overall shape of the fans, fan angle, and surface profile of the fans. Our experimental results were also compared with the theoretical predictions reported previously.

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I. INTRODUCTION

Highly nonequilibrium aggregation has been a subject of much interest in the scientific community for more than two decades. A well known and much studied case is the phenomenon of diffusion-limited aggregation.[1] In this model, particles one at a time diffuse randomly under Brownian motion toward a seed. When a particle gets near a cluster that has been formed by previous particles, it sticks to the cluster. As a result, a fractal cluster is eventually formed around the seed. Another interesting model is the ballistic aggregation (BA),[2,3] where, instead of Brownian motion, the particles shoot toward the seed randomly one at a time from all directions. Again, a cluster is formed around the seed. These models have been used to describe many real world phenomena such as colloidal aggregation of flocs and sediments.

A simplified version of ballistic aggregation restricts the direction of the incoming particles to only one direction.[4–9] In this model, the particles (parallel to each other along one direction) randomly rain down ballistically toward the seed. A particle will stick to the surface of the structure when it touches a previously added particle in the cluster. This aggregation results in the formation of a fanlike cluster. There have been many studies on the theory of the formation[6,7] of this fanlike cluster as well as on the scaling properties[5,8,9] of the fan. Although most of the studies have been performed in two dimensions, an extension to three-dimensional simulations has also been reported.[9] However, no experimental demonstrations of the formation of this fanlike structure have been published. In this paper, we report the creation of such a fanlike structure by using a simple line-of-sight evaporation technique, where atoms vertically drop down onto an array of small cylindrical seeds. This technique results in well defined fanlike clusters on the seeds. A Monte Carlo simulation with nearest-neighbor ballistic attachments followed by a diffusion mechanism has been carried out to describe the fanlike structures.

II. EXPERIMENTAL DETAILS

The cylindrical seeds are an array of tungsten pillars in the SiO_2 layer fabricated on a Si substrate. The seeds have a diameter of 150 nm and a height of 220 nm arranged in a square lattice, with a lattice constant of about 1000 nm. Scanning electron microscopy (SEM) images of these seeds are shown in Fig. 1(a). The inset is an oblique angle view of the seeds. Our experiments were conducted inside a water-

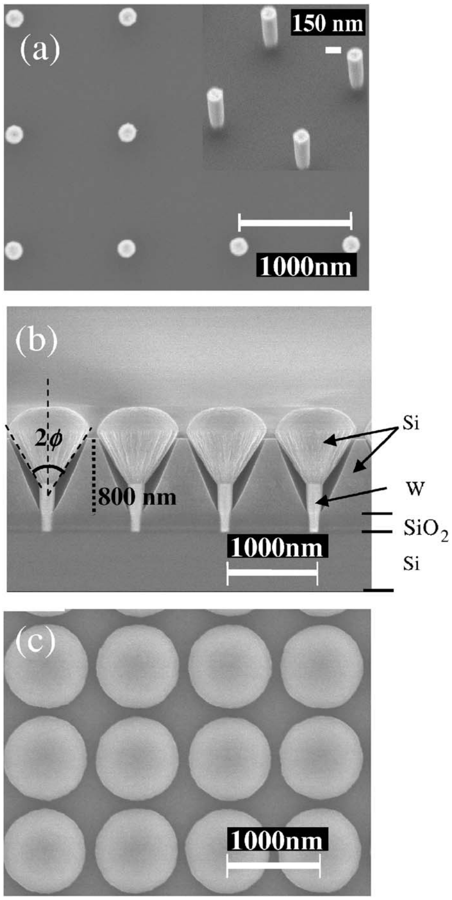

FIG. 1. (a) SEM top-view image of an array of cylindrical tungsten seeds with 150 nm diameter and 220 nm height arranged in a square lattice with 1000 nm spacing. The inset is an oblique angle view of the seeds. (b) The SEM side-view image of the ballistic fans with 800-nm-thick Si deposition. (c) The top-view image of the Si ballistic fans shown in (b).

cooled vacuum chamber with a base pressure of 9×10^{-5} Pa. We evaporated silicon (Si) at 99.9999% purity out of a cylindrical graphite crucible (with a crucible mouth of about 1 cm) using the electron bombardment method, which has been previously described.[10] The substrates containing the seeds were placed directly above the crucible at a distance of 35 cm from the mouth of the crucible. The total area of the patterned seeds is about 0.5×3 mm^2. The pressure of the chamber was maintained at $<4 \times 10^{-4}$ Pa during the deposition, and the mean free path of the vapor (>10 m) is much larger than the distance between the source and the seeds. Therefore, the Si deposition flux is highly parallel with very little angular spread. The deposition rate was 30 nm/min, and the film thickness was monitored by a quartz-crystal microbalance in the experiments. Si films were deposited at near room temperature (no intentional heating of the substrate, and the substrate temperature was less than 80 °C during the deposition). After deposition, the films were characterized by a field emission SEM system (FESEM-6350, Jeol Ltd., Tokyo, Japan).

Figures 1(b) and 1(c) show the side-view and top-view images of an 800-nm-thick Si film deposited on the seeds. Well defined three-dimensional (3D) fanlike structures were formed on top of each seed. A structure with a pyramidal shape was also deposited inside the open area between the seeds. We observe that the sidewalls of the fans are straight and can be described geometrically by three-dimensional solid cones. Therefore, the width of the cones grew linearly with time. The ϕ is the polar angle measured from the central axis of the fan. The angle 2ϕ of the fans is $66.7 \pm 0.3°$ as measured from one sidewall to the other sidewall of the fans (cones). The shape of the profile of the top surface can be described by a $\cos^{1/2} \phi$ function to be explained later.

A series of computer simulations was carried out to study the evolution of fans grown on cylindrical seeds. We developed a 3D Monte Carlo simulation based on a simple cubic lattice with the dimensions of $1024 \times 1024 \times 1024$. Preoccupied sites in a square pattern were designed to mimic the array of tungsten seeds used in our experiments. Similarly, cylindrical seeds with different sizes R_0 (=24, 36, and 48 units in diameter) were used in the simulation, although the size of the seeds in simulation does not have a direct correlation with the real size of the seeds in the experiment. The separation of the nearest seeds was 256 units from center to center. The height of each seed was 80 units. The incident flux was uniformly incident normal to the surface.

Surface diffusion was included in our simulations following the diffusion model described in detail elsewhere.[11–13] In a cycle of simulation steps, one particle was launched from a random position one unit higher than the maximum height of the interface. The particle was allowed to attach to a previously deposited particle if it moved into an empty lattice point and became a nearest neighbor of a deposited particle. When a particle was added to the interface, one particle (which could be the newly added particle itself) within a certain distance was activated to diffuse into a nearby unoccupied site, provided that the move can increase the number of neighboring bonds. The diffusion was repeated until reaching a limiting number D/F, where D is the number of candidate particles for diffusion and F (=1) is the number of deposited particles during the same time interval. The diffusion strength D/F was prescribed to be 0, 20, 50, 75, 100, and 150 in our simulations. We denoted the simulation time t' as the number of total particles sent into the simulation system. The simulated structures were sampled at a constant frequency with simulation time $t' = 8 \times 10^7$ particles. The simulated films were "cleaved" through the center to form cross-section images. Figure 2 shows the cross-section view of a series of simulated profiles as a function of time for different diffusion strengths with $R_0 = 36$ units. The value of simulated width of the fan R was obtained from the top view of the fans.

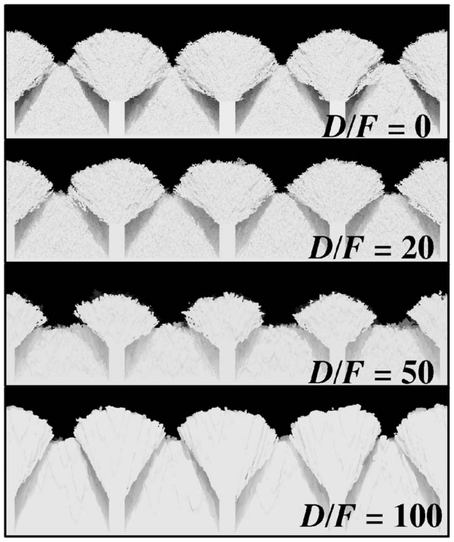

FIG. 2. Cross-section images of ballistic aggregation obtained by 3D Monte Carlo simulations with limited surface diffusion on the seeds with a diameter of $R_0 = 36$ units. The strength of diffusion D/F increases from top to bottom of the images. The simulation times for the images with $D/F = 0$, 20, 50, and 100 are 1.08×10^9, 1.28×10^9, 1.6×10^9, and 4.0×10^9 particles, respectively. Longer simulation time is necessary for the structure with stronger diffusion case to reach the same height due to the fact that the structure is more compact and has a higher density.

III. RESULTS AND DISCUSSION

We investigated the dynamic growth of the simulated width of the fanlike structures in the lateral direction, in particular, the power-law behavior of the expanding of the fans in this direction. The simulated width of the fan R was recorded as a function of time t (which is proportional to the thickness of the film). We obtained the average width R of the structures on the seeds from the top-view images. We characterized the growth behavior of R as a function of t in a power-law form,

$$R = kt^p. \tag{1}$$

The physical meaning of the exponent p is that it defines the geometrical shape of the fanlike structures. For example, a cylindrical shape has exponent $p = 0$, a hyperbolic shape is characterized by the exponent $p = \frac{1}{2}$, and a cone shape (which

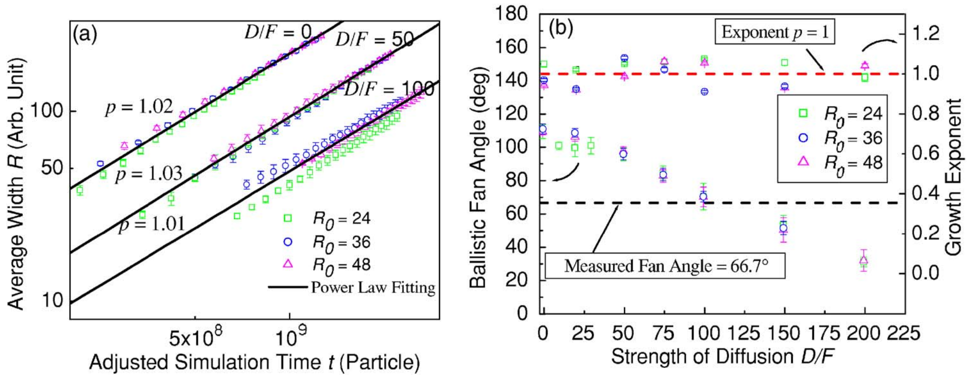

FIG. 3. (Color online) (a) The average width R of the fans is plotted in a power-law form for different sizes of the seeds R_0 and different diffusion strengths D/F. (b) The growth exponent p is plotted as a function of the diffusion strength D/F for different sizes of the seeds R_0. The simulated exponent p is a constant (≈1, indicated by the dashed line) within error for all the simulation conditions. The simulated angle of the fans changes with D/F. The experimentally measured fan angle is 66.7±0.3° and is indicated as the dashed line.

was what we observed experimentally in the present work) would have $p=1$.

In order to extract the p value more quantitatively from the simulations, we have to take into account the effect of the initial seed size. The size of the structure is equal to the size of the seeds at the beginning of the growth. The width R is therefore a function of "adjusted time" t instead of the real time t' as

$$R = kt^p, \tag{2}$$

where $t=t'+t_0$. Clearly, t_0 is an imaginary time that satisfies the constraint

$$R_0 = kt_0^p, \tag{3}$$

where R_0 is the size of the seeds. Therefore, in this power law, $R \rightarrow 0$ as $t \rightarrow 0$, which provides the possibility of comparing the simulated results with our experimental observations and with previous analytical and simulation results obtained using BA model with a point seed.

The imaginary time t_0 is not chosen arbitrarily, but in a self-consistent manner. For a set of measurements of R versus time t, we fitted the data using the power law $R=kt^p$, with a trial value of t_0 and generated the values of k and p. The condition of $R_0=kt_0^p$ was then evaluated for this trial t_0 and the corresponding k and p. If the condition cannot be satisfied, we adjusted the value of t_0 and repeated the fitting. When a satisfying t_0 is found, we plotted the width R of the simulated fanlike structure versus the adjusted time t in log-log scale as shown in Fig. 3(a). A summary of the extracted p value for different sizes of the seeds (R_0=24, 36, and 48 units) and different diffusion strengths D/F is plotted in Fig. 3(b). One can see from the plots that the growth of the fanlike structures on the seeds has a power-law behavior with the exponent $p \approx 1.0$. The width grew linearly with deposition time t. As mentioned above, the cone-shape geometry of the fans observed in our experiment is consistent with this prediction. This linear behavior is, within error, independent of the strength of diffusion and the size of seeds. Thus, we conclude that the linear growth ($p \approx 1$) of the width of the fans may be a universal characteristic of ballistic aggregation, even when diffusion is allowed.

Linear growth of the width of ballistic aggregates on a point seed has been predicted theoretically by Limaye and Amritkar based on a Poisson process[7] in a two-dimensional (2D) model. In the 2D model in Ref. 7, the average width R of the ballistic aggregates is given by

$$R = 4r^2nt, \tag{4}$$

where r is the size of the depositing particles and n is the number of particles per unit time per unit length. Even though the model is 2D, the result appears to be valid for our 3D cases. No theoretical prediction has been previously reported in the 3D case.

The morphology of the fanlike structures was further characterized by the ballistic fan angles and the surface profiles. The experimentally measured fan angle 2ϕ is 66.7±0.3° from the SEM side-view images. Previously, Porcu and Prodi predicted that the fan angle cannot be affected by the size of seeds in simulations for a large amount of particles aggregated.[14] Also, it is well established that the fan angle is larger for the 2D on-lattice simulations (about 64°) than the off-lattice counterpart (about 40°), without surface diffusion in both cases.[5–7,9,14] However, in a 3D on-lattice model, the angle is over 100° in the case of no diffusion.[9] In our 3D on-lattice Monte Carlo simulations, we observed that the fan angle depends on the strength of diffusion D/F. The ballistic fan angle obtained was around 100°–110° without surface diffusion, consistent with that reported in Ref. 9. It is also noted in Fig. 3(b) that the fan angles do not depend on the size of the seeds but depend very much on the strength of the diffusion. The angle becomes smaller as D/F increases. For $D/F \sim 100$, the angle is close to the experimental value of 66.7°. This fact indicates that the fan angle is not universal.

We studied the evolution of the surface profiles of the fanlike structures in this paper as another important geometric characteristic of the ballistic fans. The profile of the top surface of the fans deposited in experiments was mapped by measuring the height y and the distance x from the central axis of the fan for the points on the surface in the SEM side-view images. A similar method was used to obtain the profiles of the top surface in simulations at different stages of growth. We used the following approximate equation predicted by a mean-field theory[6] to fit our data:

$$H(\phi) = H_0 \cos^{1/2} \phi, \tag{5}$$

where H_0 is a time-dependent constant and ϕ is the polar angle measured from the axis of the fan. As shown in Fig. 4, the simulated surface profiles and experimental data can be fitted by this function reasonably well. Other interesting aspects of the ballistic aggregates studied in the past by computer simulations is the scaling behavior of the local density of the aggregates.[4,5,7] Unfortunately, it is not possible at this time to measure the density of the fans formed on the seeds in our experiments.

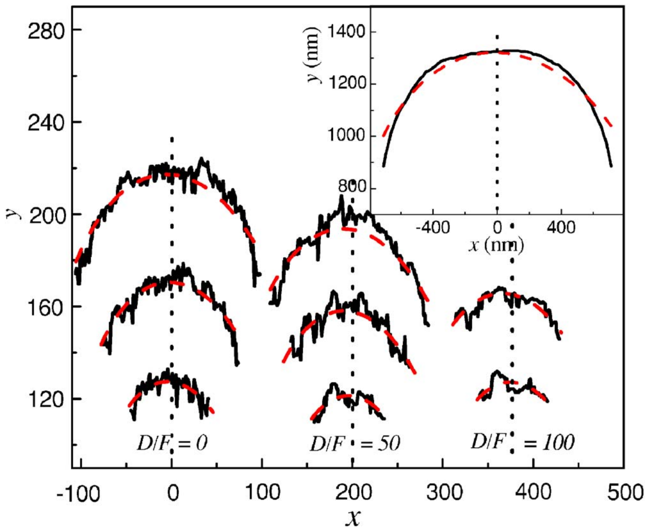

FIG. 4. (Color online) The evolution of the surface profile in simulations is plotted with $D/F=0$, 50, and 100. The dashed curves are generated by a $\cos^{1/2} \phi$ model. The inset is the surface profile of the Si fanlike structure obtained from the experiment and is fitted by the $\cos^{1/2} \phi$ model.

IV. CONCLUSION

In conclusion, we have demonstrated experimentally the fanlike growth of 3D Si structures on an array of seeds. The fans have a solid cone-shape structure. Monte Carlo simulations showed that the width of the fans evolved in time following a power-law behavior with an exponent $p \approx 1$, which is consistent with the experimentally observed cone-shape structure. This linear growth behavior is universal and predictable based on the ballistic aggregation models. In addition, the angle of the fans is independent of the size of the seeds. Our simulations suggest that surface diffusion has a prominent effect on the angle of the fans. We found that a simple $\cos^{1/2} \phi$ model predicted in the literature can be used to describe the surface profile of the fanlike structures formed in our experiments and Monte Carlo simulations fairly well.

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