Comp B: Diffusion Limited Aggregation

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Abstract. Diffusion Limited Aggregation (DLA) is a model for describing systems where growth is driven primarily by diffusion. DLA clusters are fractal objects, and computational models have been used to determine that the fractal index d_f of a 2D DLA cluster grown on a square lattice, in the limit of large cluster radius, is $d_f = 1.743 \pm 0.003$, which is in agreement with the literature value of 1.7[1]. A simulation parameter p_{stick}, the probability that a diffusing particle will stick to a DLA cluster, is introduced and it it shown that $d_f = d_f(p_{stick})$. A framework is then proposed that uses this result to link thermodynamics to the dynamics of the simulation of DLA clusters.

Preface

Although the coursework was initially presented as a C++ project, I took the liberty of rewriting the DLA algorithm in a programming language called Google Go (https://golang.org). The motivation for this is Go has been designed to accommodate systems with high concurrency. This means that instead of simulating 1 DLA cluster at a time, 1000 different clusters can be simulated simultaneously in different threads. This greatly increases time efficiency and has allowed me to achieve a significant level of statistical significance in my ensemble averages. To achieve the highest possible concurrent number of simulations, the results presented here were generated on a 32-core virtual private server. The source code for my implementation is accessible on Github: https://goo.gl/fDia6z.

1. Introduction and Simulation Method

Diffusion Limited Aggregation (DLA) is a theory that models the process of how clusters of particles form in systems where growth is driven primarily by diffusion [2]. DLA theory is applicable to systems including Hele-Shaw flow [7], electrodeposition [4] and dielectric breakdown [6].

In the DLA method used in this paper, a stationary seed particle is placed at the centre of a regular square lattice. This seed particle defines the origin of our system, as shown in Figure 1. A particle is then added to the system at a radius r_{start} from the origin. This radius is computed such that each new particle is added at a radius greater than the radius of any stationary particle in the system. Diffusion limited transport is driven by Brownian motion [5], a random process, so the new particle is made to undergo a Markovian random walk through the system. If the particle moves to a cell on the lattice that is horizontally or vertically adjacent to a stationary particle, the particle is stuck to the grid cell with a sticking probability p_{stick}. If the particle moves too far away from the cluster, as defined by a radius r_{kill}, the particle is removed from the system. This is done to increase the time efficiency of the program, as it removes the possibility of a particle moving into regions of space where the probability of sticking to the cluster in any reasonable number of further steps is extremely small. Once a particle is stuck to the cluster or killed for 'walking' too far away, a new particle is added on the start circle and the process is repeated. The simulation is terminated when the border of the cluster approaches the edge of the fixed-dimension grid or when the total number of particles in the cluster reaches a predefined limit.

In this paper, the fractal dimensions of simulated 2D DLA clusters are determined and a simulated cluster is compared with experimental results. Enquiry is also made into how fractal dimension depends on the sticking probability p_{stick}, and this is linked to the thermodynamics of physical systems that can be modelled using DLA.

2. Method

2.1. Random Walks and Ensemble Averaging

To simulate the Brownian motion of diffusing particles, DLA simulations have free particles undergo a random walk. For each iteration, the direction a particle attempts to move in is parameterized by a discrete random variable $D \sim \mathcal{U}\{0,3\} \cap \mathbb{Z}$, with each value of D corresponding to one of the particle's degrees of freedom - 2 horizontal and 2 vertical. The issue with this is that the sequence of random numbers generated by the algorithm used in the DLA simulations is only 'pseudo-random'. For a given random number generator seed, the same sequence of numbers will always be generated, which may bias the results of the simulations. To mitigate this, an ensemble of systems are simulated independently, each with a different random number generator seed. For a given measurement $\xi(t)$, an ensemble average $\overline{\xi(t)}$ is calculated by averaging measurements from each of these n systems $\{i\}_1^n$, where i represents a specific simulated system:

$$\overline{\xi(t)} = \frac{1}{n}\sum_{i=1}^{n}\xi_i(t) \tag{1}$$

2.2. Determining the Fractal Dimension of DLA Aggregates

DLA clusters are fractal objects [2]. This means their geometric structure can be described by a measure called 'fractal index', d_f. For a given DLA system, the fractal dimension is implicitly defined by [3]

$$N_c(r_{max}) = (\alpha \cdot r_{max})^{d_f} + \beta \tag{2}$$

where α, β and d_f are free parameters. Taking the natural logarithm of both sides and differentiating with respect to $\ln r_{max}$ yields the relationship

$$\frac{d(\ln r_{max})}{d(\ln N_c)} = \frac{1 + \beta/(\alpha \cdot r_{max})^{d_f}}{d_f} \tag{3}$$

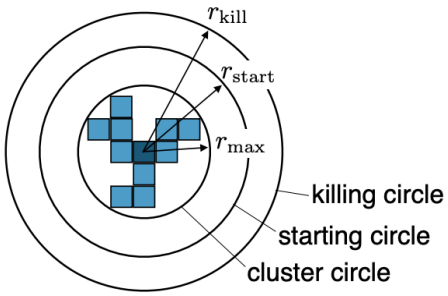
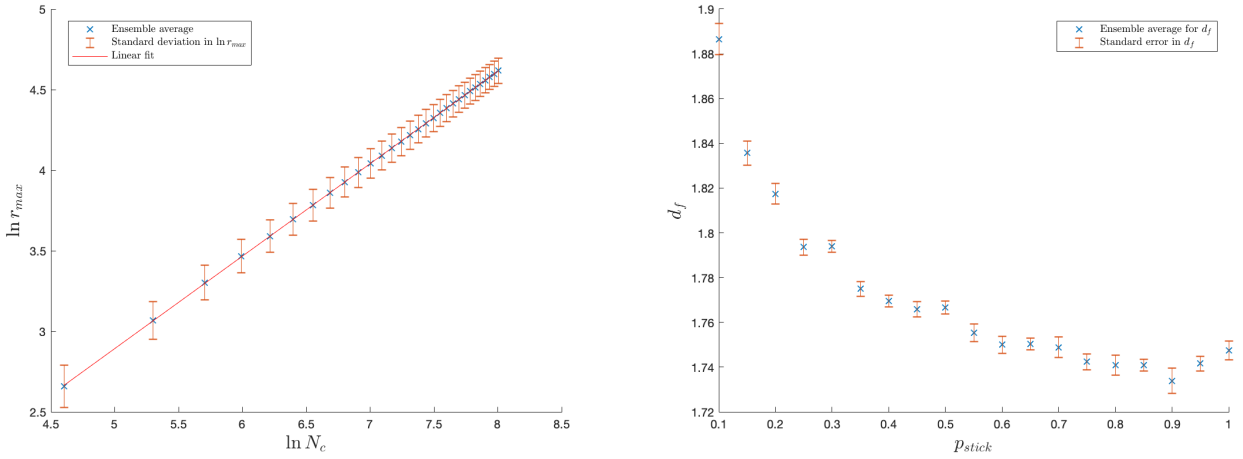


Figure 1: "Diagram of the circles used in the DLA code. The initial particle is shown in dark blue, and all circles are centred on this particle. There are three circles which indicate the approximate cluster size ('cluster circle'), the locations at which particles are introduced to the system ('starting circle'), and the points at which particles are removed from the system, to prevent them from wandering too far from the cluster ('killing circle')." Diagram and caption lifted verbatim from [3].



(a) (b)

Figure 2: (a) Natural log of cluster radius, r_{max}, as a function of the natural log of the number of particles in the cluster, N_c, for $p_{stick} = 1$. Error bars represent the standard deviation in the values of $\ln r_{max}$. (b) shows how the fractal dimension varies for a system with 2000 particles as the sticking probability is varied. For (a) and (b), data points represent an ensemble average over $n = 1000$ subsystems using equation 1.

which in the limit of large r_{max} gives

$$\lim_{r_{max}\to\infty} \frac{d(\ln r_{max})}{d(\ln N_c)} = \frac{1}{d_f} \tag{4}$$

which can be used to trivially determine the fractal dimension of a simulated DLA system.

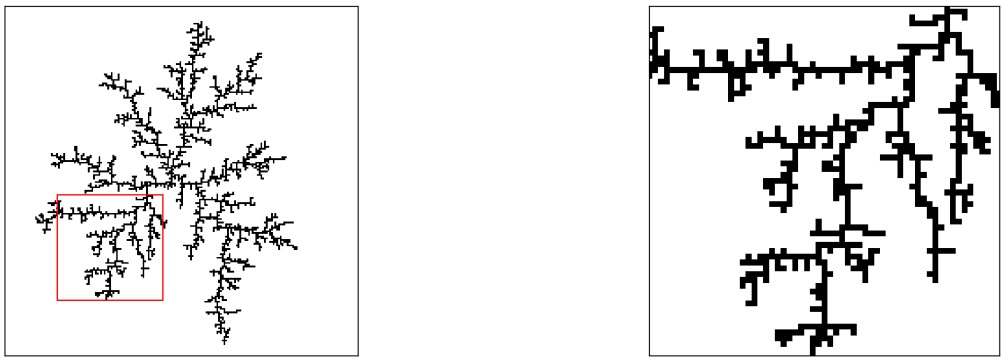
2.3. Sticking Probabilities

DLA systems can be setup so that the sticking probability p_{stick} is less than 1, indicating that a particle may sometimes collide with a cluster and recoil. This may be useful when studying systems with a high thermodynamic temperature, where free particles on average have higher kinetic energies as described by a Maxwellian distribution, for example. To achieve this, when the system determines that a particle has moved into a grid cell where there is the possibility of 'sticking', a random number p is sampled from a continuous uniform distribution $P \sim \mathcal{U}\{0, 1\}$. If $p \leq p_{stick}$ then the particle is marked as stationary and the system proceeds to add a new particle. In the case $p > p_{stick}$, the particle remains in the grid cell adjacent to the cluster but is not marked as stationary. The system then proceeds to generate a new random direction to attempt to move the particle in. If the algorithm tries to move the free particle into a grid cell that is occupied by a cluster cell, the move is rejected and new random directions are generated until a successful move is made.

3. Results

3.1. Determining the Fractal Dimensions of DLA Aggregates where $p_{stick} = 1$

Figure 2(a) shows the ensemble average of the natural log of the cluster radius r_{max} as a function of the natural log of the number of particles N_c in a DLA cluster, averaged over $n = 1000$ independent systems with different random number generator seeds and $p_{stick} = 1$. Application of equation 4 yields a fractal dimension for 2D DLA systems of $d_f = 1.743 \pm 0.003$ for systems where the sticking probability $p_{stick} = 1$.



(a) Simulated DLA Cluster (b) Magnification of lower left branch

Figure 3: Graphical representation of a simulated DLA cluster (3000 particles, $p_{stick} = 1.0$). (a) shows the entire DLA cluster and (b) shows a magnification of the bordered section in (a).

3.2. Self Similarity

Figure 3 shows graphical representations of the results of a DLA simulation with 3000 particles. Figure 3(a) shows the entire cluster and Figure 3(b) shows the bounded area from (a) magnified. Observe how all the major branching points in Figure 3(b) have exactly two child branches - one emerging downwards and one emerging to the left. This demonstrates the fractal and self-similar nature of the simulated DLA clusters.

3.3. Sticking Probabilities

Figure 2(b) shows how fractal dimension varies with sticking probability for DLA clusters with 3000 particles, with each fractal dimension data point calculated from an ensemble of $n = 1000$ independent subsystems.

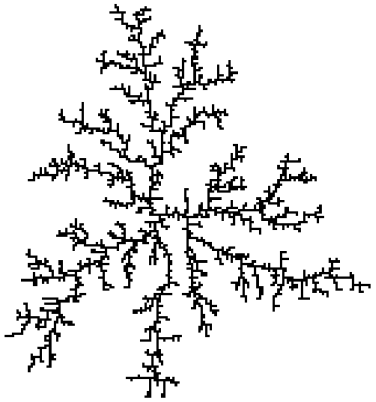
4. Discussion

4.1. Determining the Fractal Dimension of DLA Aggregates where $p_{stick} = 1$

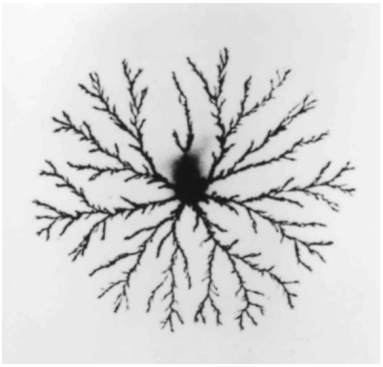
The fractal dimension of simulated 2D DLA clusters with $p_{stick} = 1$ converges on $d_f = 1.743 \pm 0.003$ for sufficiently large cluster radius, obtained by applying equation 4 to the results in Figure 2(a). This is in agreement with the literature value for 2D DLA clusters of 1.7[1]. This indicates that the simulation algorithm presented here sufficiently approximates the behaviour of physical DLA systems for cases where $p_{stick} = 1$.

4.2. Geometric Comparison with Experimental Results

Figure 4 compares a simulated DLA cluster with an experimentally produced zinc electrodeposit - a system for which diffusion is the primary transport mode, meaning DLA theory is applicable [2]. Both clusters demonstrate growth around a central attractor, with both clusters having visually similar radial branching patterns. The number of first order branches originating from the centre is greater in the experiment than in the simulated example, which could be explained by the greater surface area of the attractor in the experiment (the central electrode is not a point particle unlike the centre particle in the simulation), meaning more particles can become attached. Another consideration is that, experimentally, diffusing particles are free to attach to a cluster particle at any arbitrary angle



(a) Simulated DLA Cluster



(b) "A zinc electrodeposit produced in a thin cell" [2]

Figure 4: Comparison of a simulated DLA cluster (3000 particles, $p_{stick} = 1.0$) with an experimentally produced zinc electrodeposit as presented in [2].

$\theta \in \{\mathcal{R} \cap [0, 2\pi)\}$ (neglecting possible chemical lattice constrains), whereas in the simulations we enforce a square lattice, with free particles only attaching if they are directly horizontally or vertically adjacent to a cluster particle, thus enforcing the unnatural constraint $\theta = m\pi/2, m \in \{[0, 3] \cap \mathcal{Z}\}$. This could be mitigated by allowing diagonal attachment and could be verified by further investigating the effect, if any, of simulating DLA clusters with different lattice arrangements. However, determining if lattice arrangement affects the geometry of the produced clusters would require a more sophisticated method of quantifying the distribution and arrangement of the cluster branches, which is outside the scope of this paper. More simply, the case here could also be that, due to DLA simulations being stochastic, the same simulation setup with a different random number generator seed could produce a greater (or indeed fewer) number of branches - with the number of branches simply being 'random'.

4.3. The Effect of Sticking Probability on Fractal Dimension

Figure 2(b) indicates that fractal dimension increases as the sticking probability is reduced from 1 to 0. This makes physical sense as, for low sticking probabilities, free particles have a greater chance of reaching the 'inside' of the cluster, as opposed to immediately becoming stuck on the end of the first branch they encounter. This has the effect of increasing the number of particles in the cluster N_c for a given cluster radius r_{max}. Inspection of equation 4 confirms that increasing N_c for a given r_{max} increases the fractal dimension, as would be the effect of lowering the sticking probability.

4.4. Sticking Probabilities and Thermodynamics

The dynamical change of DLA systems as the sticking probability is changed has physical significance when linked to thermodynamics. To do so, some assumptions must be made. First, assume that the sticking probability is linearly related to the the average kinetic energy of the diffusing particles by

$$p_{stick} = \begin{cases} 1 - \dfrac{1}{\mathcal{U}_c} \cdot \overline{\mathcal{U}} & \text{for } 0 \leq \overline{\mathcal{U}} \leq \mathcal{U}_c \\ 0 & \text{for } \overline{\mathcal{U}} > \mathcal{U}_c \end{cases} \tag{5}$$

where p_{stick} is the sticking probability, $\overline{\mathcal{U}}$ is the average kinetic energy of the diffusion particles and $\mathcal{U}_c$ is a critical average kinetic energy at which point the diffusing particles are sufficiently energetic that the sticking probability becomes 0.

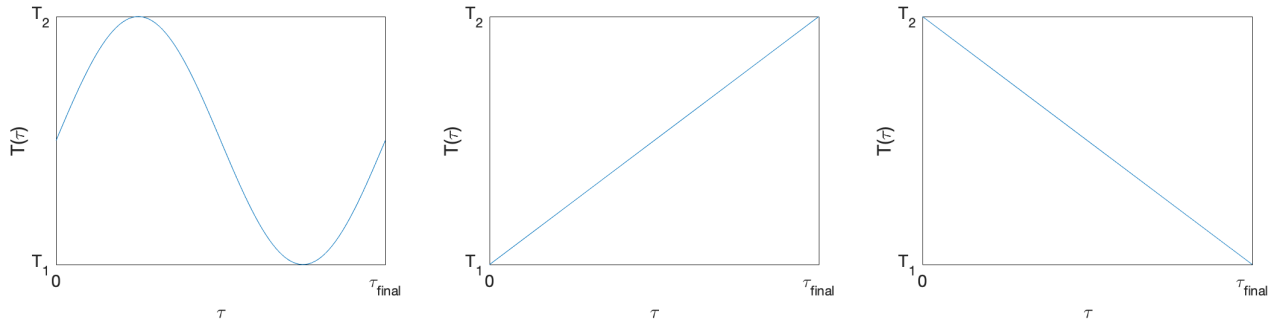


Figure 5: Possible temperature modulations for a DLA system as a function of system time τ. The first graph represents a system where the temperature is modified sinusoidally, perhaps representing in a pulse mode heating configuration. The second and third graphs represent a system being uniformly and linearly heated and cooled between two temperatures T_1 and T_2 respectively.

Further assume the kinetic theory result $T = \alpha\overline{\mathcal{U}}$ holds where T is absolute temperature and α is a real positive constant. This gives the relationship

$$p_{stick} = \begin{cases} 1 - \dfrac{1}{T_c} \cdot T & \text{for } 0 \leq T \leq T_c \\ 0 & \text{for } T > T_c \end{cases} \tag{6}$$

where T is the temperature of the DLA system and T_c corresponds to an average kinetic energy of $\mathcal{U}_c$. That is to say, at T_c, the system is so energetic that no free particles stick to the DLA cluster.

If the number of particles in the system at an instant is considered an indicator of the progress of system time, τ, then the DLA system can be setup to have a time dependent probability of sticking, $p_{stick} = p_{stick}(\tau)$. By application of equation 6, this allows the DLA system to be studied as a function of $T(\tau)$. This means we can theoretically use DLA simulations to investigate the impact of modulating temperature over time on the result of growing crystal structures, for example. Some example time dependent temperature modulations that could be of interest are shown in Figure 5. Temperature modulation as shown in the left graph could be used to model the case of pulse heating a DLA crystal structure as it grows. The second and third temperature modulations represent situations where a DLA system is gradually heated or cooled over time. Time constrains did not permit exploration of these systems, however it would be interesting to investigate if these have any affect on the geometry of the resultant crystal structures.

4.5. Suggestions for Improvement

Although seemingly effective, the implementation of the kill circle could be somewhat naive. If a particle walks outside of the kill circle it is, at present, simply terminated, and a new particle is added at random anywhere on the start circle. It may be more prudent to consider the probability of where the wandered particle is likely to reappear at a future instant of time as there could be a radial bias to the reappearance position, depending on the angle the particle wanders away from the cluster at. However, this is likely not significant as it could be expected that any radial bias would be averaged out over time because the probability distribution of the angle θ that the particle walks away from the cluster at is uniform. Notwithstanding this, a more sophisticated alternative using Green's functions and probability densities is presented in section 7.1 of "Diffusion-limited aggregation: A kinetic critical phenomenon?"[8].

Another trivial but potentially significant improvement that could be made is the addition of adaptive step sizes. In cases where the diffusing particle is far away from a cluster particle, the number of lattice points that the free particle moves per iteration could be increased, with the number of lattice points moved being proportional to the distance of the free particle from its nearest neighbour

in the cluster. This has the potential to significantly increase the time efficiency of the DLA simulation algorithm, if implemented carefully enough.

5. Conclusion

The fractal index of 2D DLA clusters with a sticking probability $p_{stick} = 1$, in the limit of large cluster radius, has been computed as $d_f = 1.743 \pm 0.003$ which is in agreement with the literature value of 1.7[1]. It has been computationally shown the fractal dimension of a 2D DLA cluster is a function of the sticking probability, with the fractal index tending to increase as the sticking probability is reduced from 1 to 0. A physical justification for this has been presented. A computational DLA cluster has been compared with a physical examples of a DLA system and possible reasons for dissimilarities have been explored. A framework for integrating thermodynamics into DLA simulations has been proposed.

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