M.Sc. Numerical Solution of Differential Equations

Numerical Modelling of Island Ripening

P resh Pre

Septe er

Acknowledgements

I would like to thank my supervisors Professor Mike Baines and Dr Paul Mulheran for all their support and helpful discussions throughout this dissertation. I would also like to thank the Department of Mathematics and my fellow students for their support and encouragement throughout this long and difficult year. Finally to my parents for all their love and support for which I am eternally grateful.

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Chapter 1

Introduction

Nanoscale devices are an upcoming advancement in the field of technology which has been a large topic of research over the past decade within academia. Nanotechevolution on a material substrate. Figure 1 shows the basic procedure that takes place in the evolution process.

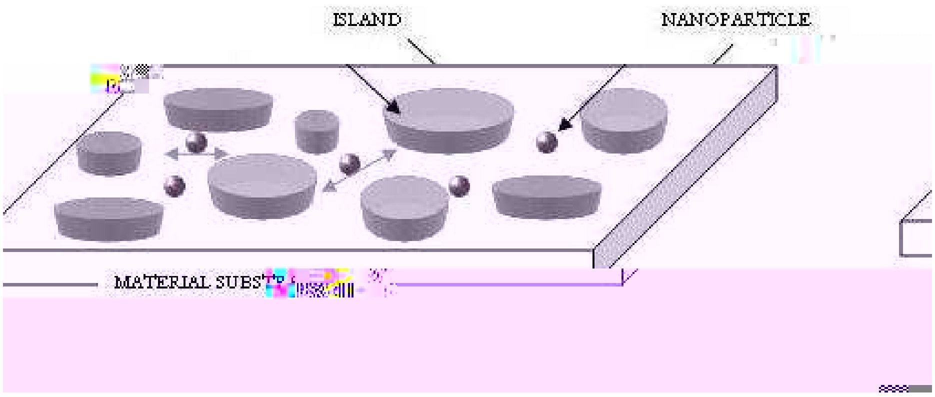


Figure 1.1: *Island evolution on a material substrate*

An island is a group of nanoparticles, Figure 1, considered as one entity and varying in size. Once these islands have formed and with no addition of nanoparticles added to the system, i.e a set amount of material on the surface, we look to see how the islands evolve over time. The evolution process involves particles moving about from different islands where the number of islands starts to decrease and at some point will slow down enough that we can say that it is reached a quasi-steady state. Note that the ripening process does not stop and islands will continue to disappear where all depends on the temperature of the system although we will not consider this variable in our calculations. The movement is due to different types of interactions [3] that can be driving the evolution, of which part of the research in this area is trying to study.

In this project we look at two properties of island evolution, the growth rate equation for an island and the distribution function. The growth rate equation describes how an island can possibly evolve and the distribution function is a measure of islands over the whole surface. Firstly, we look at the origin of the growth rate equation.

1.1 Origin of the Growth Rate Equation

1.1.1 Lifshitz and Slyozov

The equations that we look at have their origin in a paper written by Lifshitz and Slyozov [1] in 1961. They look at the diffusion effects of the precipitate formed in a supersaturated solid solution. These diffusion effects bring about the formation of grains of a new phase in the solution. The grains arise due to the supersaturated part of the solution and then grow due to the coalescence effects brought about

volume distribution of grains in the solution. The volume distribution function is an unknown quantity that we wish to find and is related to the growth law via the continuity equation,

$$\frac{\partial f(\rho^3, t)}{\partial t} + \frac{\partial}{\partial \rho^3}(\dot{V} f(\rho^3, t)) = 0. \tag{1.7}$$

The first term is the time rate at which the distribution increases and the second term tells us the accumulation of material due to the grain growing or dissolving in the solution. Note that the equation is essentially one-dimensional although the quantities considered are three-dimensional in nature.

Conservation

A conservation property must hold for this system. As a grain grows the amount of supersaturation must reduce to compensate for the growth. The grain grows due to the over saturation of the solution. The total initial supersaturation of the solution is

$$Q_0 = \Delta_0 + q_0, \tag{1.8}$$

where the initial supersaturation Δ_0 is as before and also a term q_0 which allows for the initial volume of material already in the grains. This term is quantified through

$$q_0 = \frac{4}{3}\pi R_{c0}^3 \int_0^\infty f \rho^3 d\rho^3 \tag{1.9}$$

which is the volume of a grain multiplied by the first moment of the volume distribution function. Also as we already know, the number density, n, of grains (number of grains per unit volume) is represented by the area under the curve which is the normalised zeroth moment

$$n = \int_0^\infty f \, d\rho^3. \tag{1.10}$$

Lifshitz and Slyozov then use the normalisation to unit volume to relate a one-dimensional distribution function $F(\rho, t)$ to the three-dimensional volume distribution function via

$$F(\rho, t) d\rho = f(\rho^3, t) d\rho^3. \tag{1.11}$$

1.1.2 Hillert

Further to Lifshitz and Slyozov [1], Hillert [2] approached the problem of grain growth from a di�erent view point but ultimately ending up with a similar growth equation. This was done deliberately so that the method of Lifshitz and Slyozov could be implemented when coming to solve the continuity equation. The grain growth equation was

$$\frac{dR^2}{dt} = 2\alpha M\sigma\left(\frac{R}{R_c} - 1\right), \tag{1.14}$$

where M, α and σ are parameters controlling how and when grains come together. Note here that the growth equation describes the rate of change of grain size rather than grain volume, as Hillert found this to be easier to study theoretically. The scaled distribution function, $P(u)$, then comes out to be

$$P(u) = (2e)^2\frac{2u}{(2-u)^4}\exp\left\{\frac{-4}{2-u}\right\}, \tag{1.15}$$

where $u = R/R_c$

The Monte Carlo data look at two types of evolution; the pedophagous effect (PE) and the non-pedophagous effect (NPE). The PE effect is when a particle escapes from an island but the same island is then able to capture it back, thus enabling it to absorb its own offspring, while the NPE is when this is not allowed to happen. The results found were that the Hillert growth law in the 2-D form

$$\dot{s} = \frac{ds}{dt} = \frac{r}{\bar{r}(t)} - 1, \qquad (1)$$

asymptotic nature of the solution. The conclusion reached is that a spatial order of islands arise due to the PE but not in the NPE case. The growth rate equations of Hillert and Tarr and Mulheran are the ones we go on to study.

Chapter 2

Asymptotic Analysis

2.1 Tarr and Mulheran

The quasi-steady state solution as $t \to \infty$, i.e the scaled distribution function, has been the preferred choice for solving the continuity equation since an analytical number density of islands, $N(t)$. Note that the volume of material, Ω should

Tarr's Law then takes a special case and chose $f(0) = f_0 = c/k = 1$ where $f(0)$ comes from

$$\dot{N} = -k\frac{\Omega}{ct^2} = \lim_{s\to 0}\left(\frac{ds}{dt}F(s,t)\right) = -k\frac{\Omega}{c^2t^2}f(0) \tag{2.7}$$

which describes how islands are able to disappear as $s \to 0$ with a rearrangement giving the required special case. Thus, the ODE reduces to

$$-f - f' = 0 \tag{2.8}$$

which has the solution

$$f(v) = \exp(-v). \tag{2.9}$$

However, other solutions may exist by solving the original ODE 2.6 with a general $c/k = f_0$. This 0

$$f(v) = f_0[1 - (1 - f_0)v]^{\frac{1-2f_0}{f_0-1}}. \tag{2.13}$$

So Tarr's Law [4] finds two solutions to the continuity equation of which the special case seems to be the solution of choice experimentally and the one that also should

$$\frac{\partial F(s,t)}{\partial s} = \frac{\Omega}{c^3 t^3} f'(v), \tag{2.18}$$

from equation 2.4. Now, using equation 2.18 the complete size derivative in the continuity equation we find

$$\frac{\partial}{\partial s}\Big[(v^{1\ 2} - 1)F(s,t)\Big] = \frac{\Omega}{c^3 t^2}\Big[(v^{1\ 2} - 1)f'(v) + \frac{f(v)}{2v^{1\ 2}}\Big]. \tag{2.19}$$

Substituting equations 2.17 and 2.19 into the continuity equation 2.16 we find the ODE

$$f[1 - 4cv^{1\ 2}] + f'[2(v - v^{1\ 2}) - 2cv^{3\ 2}] = 0, \tag{2.20}$$

where separation of variables gives

$$\int \frac{df}{f} = \int \frac{1 - 4cv^{1\ 2}}{[2(v - v^{1\ 2}) - 2cv^{3\ 2}]}\, dv. \tag{2.21}$$

Theef

F

c

and if $4c > 1$

$$f(v) = A(cv - v^{1\ 2} + 1)^{-2} \exp\left\{ \frac{-2}{\sqrt{4c-1}} \arctan\left[\frac{2cv^{1\ 2} + 1}{\sqrt{4c-1}} \right] \right\} \qquad (2.25)$$

where in both cases we have converted back to the scaled island size, v, with A integration constant is dependent on the allowed values for v. We can also find a solution when 4c d 1

By setting the term in the square brackets to zero the characteristics are given by the growth rate equation 2.1 where the constant k has been scaled into the time derivative. The function u is not constant on the characteristics due to the total derivative of $u(s,t)$ being non zero

$$\frac{du}{dt} = -\frac{u}{\bar{s}}. \tag{2.35}$$

The characteristic equation is analytically solvable for the quasi-steady state of the system when we replace $\bar{s}$ with t, in this case taking $c = 1$ for simplicity. The growth rate equation becomes

$$\frac{ds}{dt} = \frac{s}{t} - 1 \tag{2.36}$$

which can be solved using the integrating factor t^{-1} to give

$$\frac{d}{dt}(t^{-1}s) = -t^{-1}. \tag{2.37}$$

Integrating we find

$$s = -t\ln t + Bt, \tag{2.38}$$

where B is a constant of integration.

However, since u is not constant on the characteristics, i.e $\dot{u} \neq 0$, we can solve equation 2.35 as well, again with $\bar{s} = t$, which tells us what happens on the characteristics. This is trivial coming out to be

$$u = At^{-1}, \tag{2.39}$$

where A is a constant of integration.

Chapter 3

Numerical Schemes

$\mathbf{u}^n$

to give

$$u_{tt} = \frac{\dot{\bar{s}}}{(\bar{s})^2}u + \frac{\dot{\bar{s}}}{(\bar{s})^2}su + \frac{1}{\bar{s}}((\dot{s}u)_s) + \dot{s}(\dot{s}u_{ss} + \frac{2\dot{s}}{\bar{s}}u_s). \tag{3.12}$$

Finally substituting the above into 3.5 we obtain

$$\begin{aligned} u(s, t + \Delta t) &= u(s, t) - \Delta t(\dot{s}u)_s \\ &+ \frac{(\Delta t)^2}{2}\left[\frac{\dot{\bar{s}}}{(\bar{s})^2}u + \frac{\dot{\bar{s}}}{(\bar{s})^2}su_s + \frac{1}{\bar{s}}((\dot{s}u)_s) + \dot{s}(\dot{s}u_{ss} + \frac{2\dot{s}}{\bar{s}}u_s)\right]. \end{aligned} \tag{3.13}$$

However $\dot{\bar{s}}$ is non trivial and is calculated from the initial $\bar{s}$ value given by equation 2.2, using the product rule to give

$$\dot{\bar{s}} = \frac{\int_0^\infty suds}{(\int_0^\infty uds)^2}\int_0^\infty (\dot{s}u)_s ds - \frac{\int_0^\infty s(\dot{s}u)_s ds}{\int_0^\infty uds}. \tag{3.14}$$

Calculating the integrals, $\dot{\bar{s}}$ becomes in computable form

$$\dot{\bar{s}} = \frac{\int_0^N suds}{(\int_0^N uds)^2}[\dot{s}u]_0^N - \frac{1}{\int_0^N uds}\left([\dot{s}su]_0^N - \int_0^N (\dot{s}u)ds\right), \tag{3.15}$$

where the integration limits are now on the finite region [0,N] so as to be computable. The scheme is thus achieved, as in the L-W schemes, by discretising 3.13 using central differences, giving

$$\begin{aligned} u_j^{n+1} = u_j^n - \Delta t&\left[\frac{(\dot{s}u)_{j+1}^n - (\dot{s}u)_{j-1}^n}{2\Delta s}\right] \\ + \frac{(\Delta t)^2}{2}&\left\{\frac{\dot{\bar{s}}}{(\bar{s})^2}\left[u_j^n + s_j^n\left(\frac{u_{j+1}^n - u_{j-1}^n}{2\Delta s}\right)\right]\right. \\ &+ \frac{1}{\bar{s}}\left[\frac{(\dot{s}u)_{j+1}^n - (\dot{s}u)_{j-1}^n}{2\Delta s}\right] \\ &+ \frac{2\dot{s}_j}{\bar{s}}\left(\frac{u_{j+1}^n - u_{j-1}^n}{2\Delta s}\right) \\ &\left. + \dot{s}_j^2\left(\frac{u_{j+1}^n - 2u_j^n + u_{j-1}^n}{(\Delta s)^2}\right)\right\}. \end{aligned} \tag{3.16}$$

This is longer than the standard L-W scheme but is still computable.

3.2.1 The CFL Stability Condition

We cannot perform the usual fourier stability analysis for this scheme so we look to the CFL stability condition which is a necessary condition for stability for numerical schemes of this sort [5, 6]. The stability condition we require is

$$\left|\dot{s}\frac{\Delta t}{\Delta s}\right| \leq 1, \tag{3.17}$$

where

$$\dot{s} = \frac{s}{\bar{s}} - 1. \tag{3.18}$$

Initially when s is large, depending on what domain size is taken, the value of $\dot{s}$ can be very large. Therefore, if the ratio $\Delta t/\Delta s$ is not small enough the stability condition can be violated. By choosing a smaller time step, Δt, we can avoid this problem in the case of large s but at small s the condition is always satisfied. Note that initially $\bar{s}$ is 1 but increases as time evolves which keeps the stability condition satisfied from the initial time step.

3.3 Conservation

Numerically we can lose conservation of material if the correct boundary conditions are not implemented but for this system we can always check the property

$$\int_0^\infty su(s,t)ds = 1 \tag{3.19}$$

which tells us that the volume of material is constant. Notice that the integral is equal to 1, CFL s,1

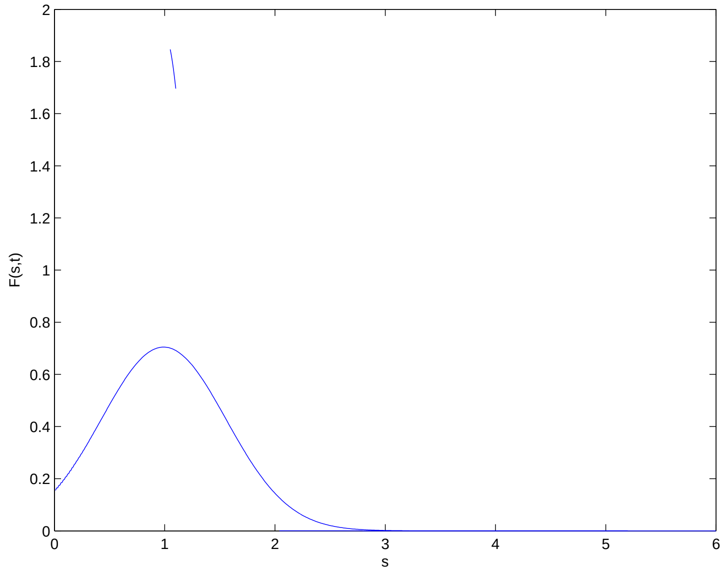
Chapter 4

Numerical Results

4.1 Tarr and Mulheran Growth Law: Gaussian

We now have a second order L-W like numerical scheme that we can use to solve the continuity equation 2.30. The conditions we start with are a Gaussian initial condition with peak centered at $\bar{s}$

Figure 4.1 shows what happens as we evolve to t = 1 where the initial Gaussian condition is also shown for comparison.



$Time$	$N(t)$	**Ω**	**s̄**
0.0000000	**0.99999902**	**1.0000000**	**1.0000010**

Figure 4.2 shows the solution obtained at $t = 5$. Note that the initial condition has been omitted so that the solution can be seen more clearly. Here the solution has started to look more like an exponential solution, as we expect, and $N(t)$ has also reduced in size from Table 4.1. There is a slight problem with the scheme though. In Figure 4.2 there are very small oscillations that are just about visible at about $s = 2$ which is because of the central difference nature of the numerical scheme [5] where we know the L-W scheme generates oscillations.

However, they do not cause any problems since the linear relationship of $\bar{s} \propto t$ has been achieved in Figure 4.3 so now we can change to the solution from the characteristic equations.

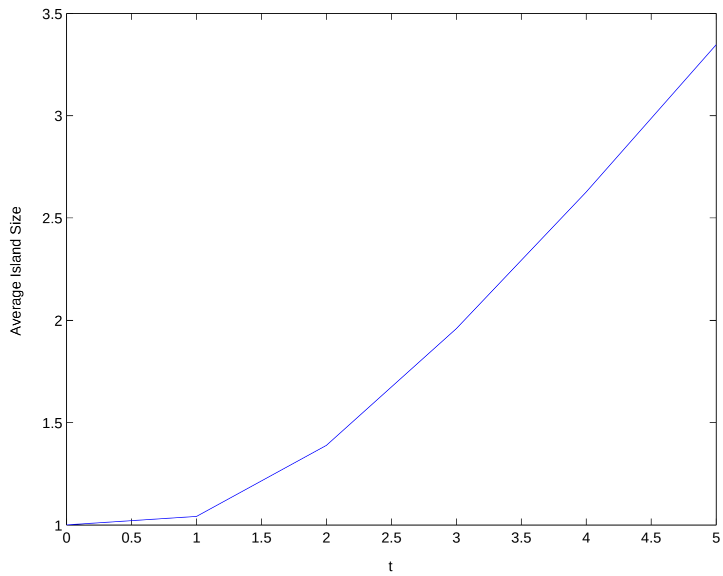


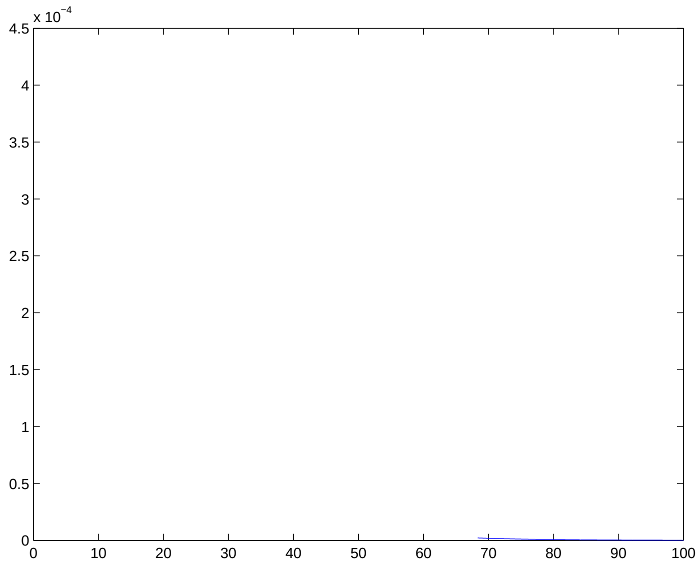
Figure 4.3: *This graph shows the linear relationship* $\bar{s} \propto t$ *has been attained by evolving to a time of* $t = 5$

Before we calculate this we can make another comparison to check our numerical results by converting to the scaled variables, f and

$$v = \frac{N(t)}{\Omega} s, \tag{4.5}$$

where $N(t)$ and Ω represent the integrals defined above. Figure 4.4 shows the scaled distribution function at $t = 5$ and also at $t = 10$ and $t = 20$ to show that the exponential solution found by the scaling solution is sl by rearranging the characteristic equations

$$u = At$$


x 10^-4
4.5
4
3.5
3
2.5
2
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1
0.5
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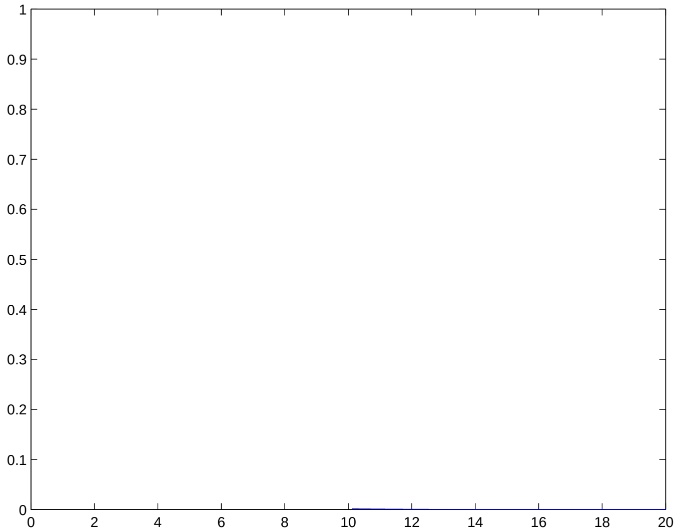
80
90
100


1
0.9
0.8
0.7
0.6
0.5
0.4
0.3
0.2
0.1
0
0
2
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8
10
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14
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18
20

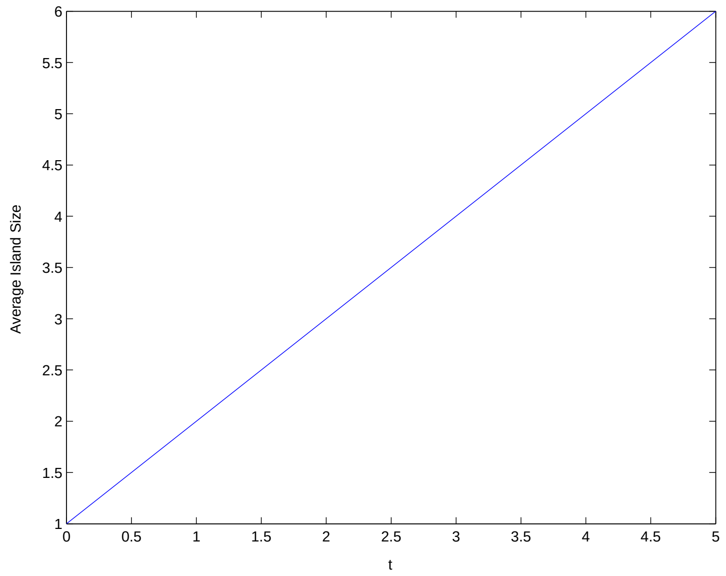
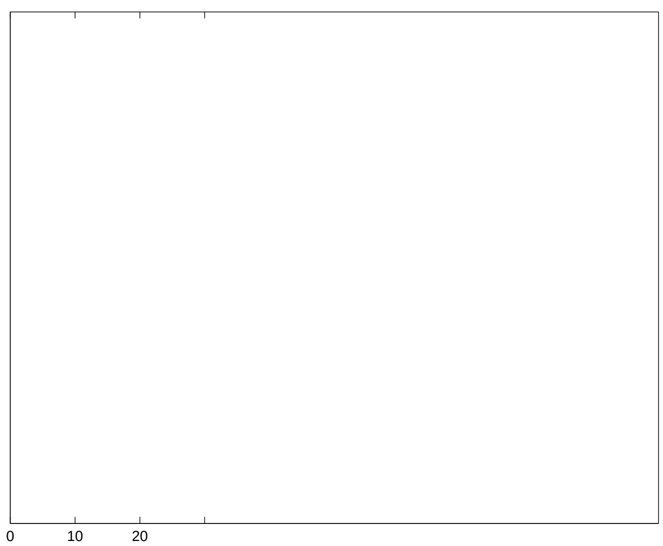
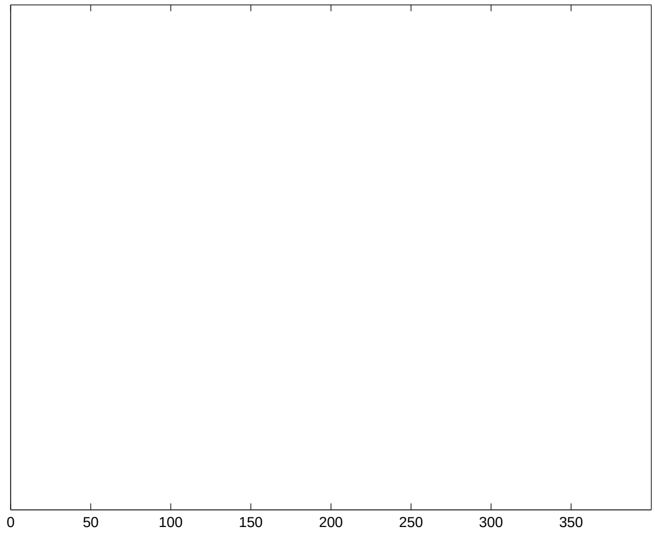


Figure 4.9: *This graph shows the linear relationship* $\bar{s} \propto t$ *has been attained by evolving to a time of* $t = 5$




0
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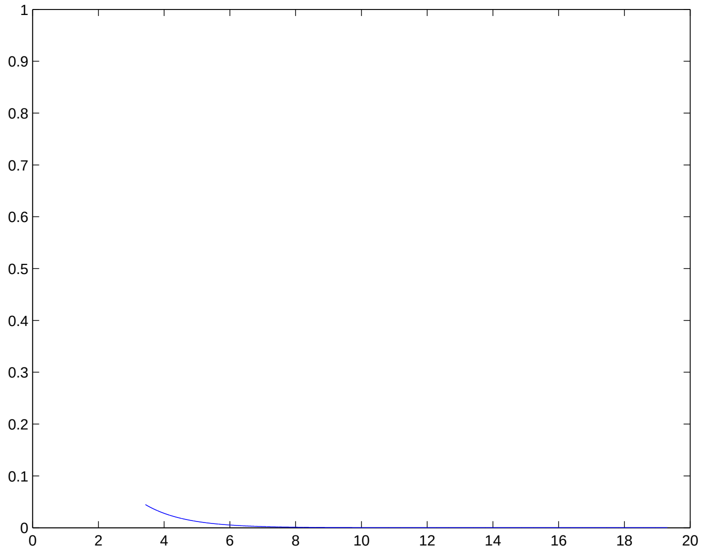
200
250
300
350



1
0.9
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0.7
0.6
0.5
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12
14
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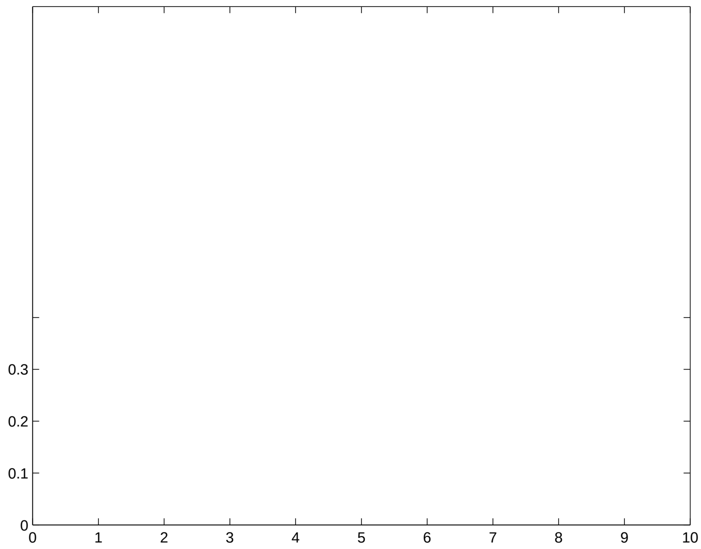
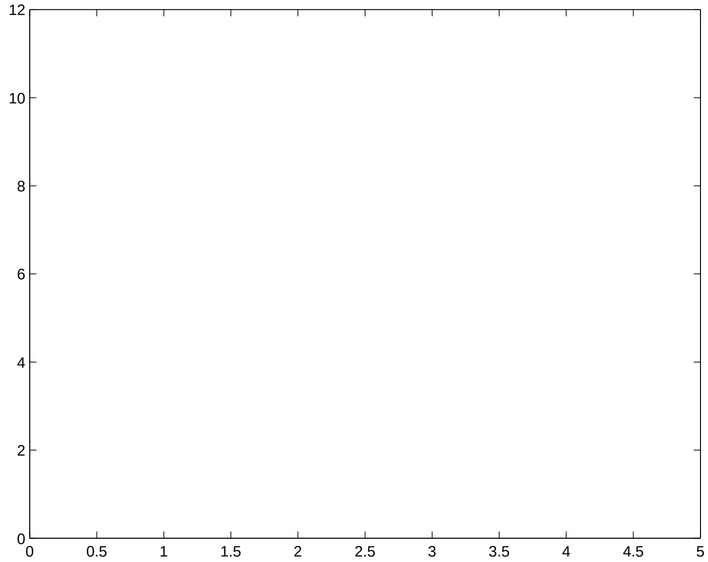
18
20


0.3
0.2
0.1
0
0
1
2
3
4
5
6
7
8
9
10

$Time$	$N(t)$	**Ω**	**s̅**
0.0000000	**0.50002300**	**0.49799765**	**0.99594947**
0.9999975	**0.16687689**	**0.49600964**	**2.97230861**
1.9999949	**0.10030785**	**0.49402960**	**4.92513360**
2.9999924	**7.1781E-02**	**0.49205745**	**6.85496147**
3.9999899	**5.5932E-02**	**0.49009317**	**8.76219618**
4.9999874	**4.5846E-02**	**0.48813672**	**10.6472200**

Table 4.3: *Table of Momens()()-(M)-(o)-()-()Tf()()()()refref*


12
10
8
6
4
2
0
0
0.5
1
1.5
2
2.5
3
3.5
4
4.5
5

Chapter 5

Conclusions and Further Work

Taking the Tarr and Mulheran growth rate equation we found that the assumption where the average island size grows linearly with time in the

the Tarr and Mulheran growth rate equation. A general power could be used here along with a numerical scheme that can sufficiently deal with the continuity equation produced.

Appendix A

Conservation Property

There is a conservation property we can check to ensure that w

where we can take out the time derivative to yield the conserving property A.1 in the first term

$$\frac{d}{dt}\int_0^\infty \left(sF(s,t)\right)ds + \int_0^\infty s\frac{\partial}{\partial s}\left(\dot{s}F(s,t)\right)ds = 0. \tag{A.5}$$

The first integral on the left is the conservation property which we have said should be constant. Therefore, the conservation property holds if

$$\int_0^\infty s\frac{\partial}{\partial s}\left(\dot{s}F(s,t)\right)ds = 0. \tag{A.6}$$

We can integrate by parts to give

$$\left[s\dot{s}F(s,t)\right]_0^\infty - \int_0^\infty \dot{s}F(s,t)ds \tag{A.7}$$

and substituting $\dot{s}$ with the growth rate equation 2.1 we get

$$\left[s\dot{s}F\right]_0^\infty - \int_0^\infty \left(\frac{sF}{\bar{s}} - F\right)ds \tag{A.8}$$

which can be written as

$$\left[s\dot{s}F\right]_0^\infty - \frac{1}{\bar{s}}\int_0^\infty sF\,ds - \int_0^\infty F\,ds, \tag{A.9}$$

since $\bar{s}(t)$ is only a function of time. Substituting the integral form of $\bar{s}(t)$ 2.2 we

This term is zero since we know that $F(\infty, t)$ tends to zero quicker than any other term. Hence from A.5

$$\frac{d}{dt}\int_0^\infty \Big(sF(s,t)\Big)ds = 0 \tag{A.12}$$

and this conservation property is valid.

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