Pattern Selection in a Phase Field Model for Directional Solidification

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A symmetric phase field model is used to study wavelength selection in two dimensions. We study the problem on a finite system by a two-pronged attack. First we construct an action and minimizing this we obtain the most probable configuration of the system which we identify with the selected stationary state. The minimization is constrained by the stationary solutions of stochastic evolution equations and is done numerically. Secondly, additional support for this selected state is obtained from straightforward simulations of the dynamics from a variety of initial states.

The problem of pattern selection is to predict which pattern, out of a number of possible stationary patterns, will be selected under given experimental conditions. This problem has been of great interest for many years in many fields such as biology, chemistry, engineering and physics [1] where a large variety of reproducible patterns can be formed under various external conditions which drive the system away from thermal and mechanical equilibrium. Important examples of pattern selection are directional solidification and eutectic growth where the interface between the ordered and disordered phases can take a periodic cellular pattern. From the experimental point of view, the interface seems to have a well defined periodicity in both directed one phase solidification [2–4] and in directed eutectic solidification [5, 6]. On the theoretical side, there is much disagreement with some workers claiming that a unique wavelength is selected [7–11] while others say that the wavelength of the final pattern is accidental [12, 13]. A large scale simulation on a noisy Swift Hohenberg equation [14] which models fluid convection near onset has been done [15] to study the selection of periodic patterns. For this system, a potential $\mathcal{F}_{SH}$ exists and minimizing this gives the unique selected stationary pattern for any initial state.

We study a related pattern selection problem in a phase field model with additive stochastic noise developed for growth processes such as single phase directional solidification [16–18] driven out of equilibrium by an external moving temperature gradient. This causes invasion of a disordered by an ordered phase with non potential dynamics. Since we are unable to solve the problem analytically, we use numerical computations which limits us to rather small system sizes. However, we believe that this is sufficient to demonstrate our conjecture of a unique selected state. The equilibrium system is described by a free energy functional $\mathcal{F}(c,\psi)$ [16] where

$$\begin{aligned}\mathcal{F}(c,\psi) &= \int d\mathbf{r}\Big(\frac{K_\psi}{2}(\nabla\psi)^2+\frac{K_c}{2}(\nabla c)^2+f(c,\psi)\Big)\\ f(c,\psi) &= -\frac{1}{2}\psi^2+\frac{1}{4}\psi^4+\Delta T h(\psi)+\frac{1}{2}\gamma\Delta\psi\Big(c+\frac{h(\psi)}{\Delta\psi}\Big)^2\\ h(\psi) &= \frac{15}{8}\Big(\psi-\frac{2}{3}\psi^3+\frac{1}{5}\psi^5\Big).\end{aligned} \tag{1}$$

Here $h(\psi)$ an odd function of ψ and $h'(\pm1)=0$ so that the equilibrium values $\psi_{eq}=\pm1$ are independent of c [19] and $\Delta\psi=2$. Since the interface between the ordered $\Delta T<0$ and disordered $\Delta T>0$ phases has width $\xi=\mathcal{O}(1)$, we define its position by $\psi(x,z)=0$.

To impose motion of the interface, we take $\Delta T(\mathbf{r},t)=\Delta T(z-vt)$ where $\mathbf{v}=v\hat{\mathbf{z}}$ is the externally imposed pulling velocity. Stationary states can exist only in the frame moving with the interface when $\Delta T(\mathbf{r},t)=\Delta T(z)$. The simplest possible dynamics consistent with the macroscopic conservation law for c are Langevin equations [20]

$$\begin{aligned}\dot\psi=\frac{\partial\psi}{\partial t} &= -\Gamma_\psi\frac{\delta\mathcal{F}}{\delta\psi}+v\frac{\partial\psi}{\partial z}+\eta_\psi\equiv\mathcal{G}_\psi+\eta_\psi\\ \dot c=\frac{\partial c}{\partial t} &= \Gamma_c\nabla^2\frac{\delta\mathcal{F}}{\delta c}+v\frac{\partial c}{\partial z}+\eta_c\equiv\mathcal{G}_c+\eta_c\end{aligned} \tag{2}$$

which define $\dot\psi$, $\dot c$, $\mathcal{G}_\psi(c,\psi)$ and $\mathcal{G}_c(c,\psi)$. Complications such as fluid flow are not considered. The stochastic noises $\eta_\psi(\mathbf{r},t)$ and $\eta_c(\mathbf{r},t)$ have probability distributions

$$\mathcal{P}(\eta_c)\propto\exp\Big(-\frac{1}{4\Gamma_c kT}\int dtd\mathbf{r}d\mathbf{r}'\eta_c(\mathbf{r},t)G(\mathbf{r},\mathbf{r}')\eta_c(\mathbf{r}',t)\Big)$$

$$\mathcal{P}(\eta_\psi) \propto \exp\Big(-\frac{1}{4\Gamma_\psi kT}\int dt d\mathbf{r}(\eta_\psi(\mathbf{r},t))^2\Big) \tag{3}$$

where T is a temperature and k is Boltzmann's constant. $G(\mathbf{r},\mathbf{r}')$ obeys $\nabla^2 G(\mathbf{r},\mathbf{r}') = -\delta(\mathbf{r}-\mathbf{r}')$ with appropriate boundary conditions and $G^{-1}(\mathbf{r},\mathbf{r}') = -\nabla^2\delta(\mathbf{r}-\mathbf{r}')$ so

$$\begin{aligned}\langle \eta_\psi(\mathbf{r},t)\eta_\psi(\mathbf{r}',t')\rangle &= 2\Gamma_\psi kT\delta(\mathbf{r}-\mathbf{r}')\delta(t-t') \\ \langle \eta_c(\mathbf{r},t)\eta_c(\mathbf{r}',t')\rangle &= 2\Gamma_c kTG^{-1}(\mathbf{r},\mathbf{r}')\delta(t-t').\end{aligned} \tag{4}$$

The form of Eq. (4) is required by the dynamics of the nonconserved field ψ and the conserved field c [20]. We assume η_ψ and η_c obey a fluctuation dissipation theorem but this restriction is not necessary and makes no qualitative difference. The exact form of the Green's function $G(\mathbf{r},\mathbf{r}')$ in Eq. (3) is needed for numerical computations.

We formulate the problem in terms of an action $\mathcal{S}(\psi,c)$ whose minimum gives the most probable or selected state. The joint probability distribution $\mathcal{P}(\psi,c)$ is obtained from the generating function

$$\mathcal{Z} = \int \mathcal{D}\eta_\psi\mathcal{D}\eta_c\mathcal{P}(\eta_\psi)\mathcal{P}(\eta_c) = \int \mathcal{D}\psi\mathcal{D}c\exp\Big(-\mathcal{S}(\psi,c)\Big) \tag{5}$$

using Eqs. (2,3). Note that the Jacobian for the variable change $(\eta_\psi,\eta_c)\to(\psi,c)$ is unity [21]. The configuration (ψ,c) minimizing $\mathcal{S}(\psi,c)$ is the most probable state at least for weak noise [22, 23].

The explicit expression for the action is

$$\mathcal{S}(\psi,c) = \int d\mathbf{r}dt\frac{\Big(\dot\psi(\mathbf{r},t)-\mathcal{G}_\psi(\mathbf{r},t)\Big)^2}{4kT\Gamma_\psi} + \int dtd\mathbf{r}d\mathbf{r}'\frac{\Big(\dot c(\mathbf{r},t)-\mathcal{G}_c(\mathbf{r},t)\Big)G(\mathbf{r},\mathbf{r}')\Big(\dot c(\mathbf{r}',t)-\mathcal{G}_c(\mathbf{r}',t)\Big)}{4kT\Gamma_c} \tag{6}$$

where $\mathcal{G}_\psi$ and $\mathcal{G}_c$ are defined in Eq. (2) and $G(\mathbf{r},\mathbf{r}')$ is the Green's function. Note that this quadratic form for $\mathcal{S}(\psi,c)$ is very similar to the actions proposed previously [11, 24, 25]. We are interested in stationary patterns at very large times t. A reasonable assumption is to argue that the pattern becomes very close to the selected stationary pattern at some large but finite t_0 and remains in this stationary configuration $\psi(\mathbf{r}), c(\mathbf{r})$ for $t > t_0$ when $\dot\psi = 0 = \dot c$. Then, the action is dominated by the infinitely long time interval $\mathcal{T} = t - t_0 \to \infty$ and becomes

$$\frac{4kT\mathcal{S}(\psi,c)}{\mathcal{T}} = \frac{1}{\Gamma_c}\int d\mathbf{r}d\mathbf{r}'\mathcal{G}_c(\mathbf{r})G(\mathbf{r},\mathbf{r}')\mathcal{G}_c(\mathbf{r}') + \frac{1}{\Gamma_\psi}\int d\mathbf{r}\Big(\mathcal{G}_\psi(\mathbf{r})\Big)^2 + \mathcal{O}(\mathcal{T}^{-1}) \tag{7}$$

As discussed below Eq. (5), the configuration $\psi(\mathbf{r})$, $c(\mathbf{r})$ which minimizes the action $\mathcal{S}(\psi,c)$ in Eq. (7) maximizes the probability $\mathcal{P}(\psi,c)$. This is the selected stationary state if dynamical paths exist between any intermediate state and the selected state in analogy with the ergodic theorem. This is almost certainly true but is very hard to verify by simulations as metastable states in the Eckhaus band [26] have very long lifetimes with weak noise even for our very small systems. This is similar to metastable states separated from the equilibrium state by large free energy barriers. The essential difference in the case studied here is that the dynamics of Eq. (2) is non potential while the approach to equilibrium is governed by potential dynamics [15]. A naive minimization of the action $\mathcal{S}(\psi,c)$ of Eq. (6) yields the deterministic evolution equations which are incorrect with noise. The minimization must be done subject to the constraints that $\psi(\mathbf{r},t)$ and $c(\mathbf{r},t)$ are solutions of Eq. (2) which we perform numerically. In principle, this can be done analytically in the Martin-Siggia-Rose formalism [27], as used in studies of the KPZ system [28], at the expense of introducing two auxiliary Lagrange multiplier fields $\tilde\psi(\mathbf{r},t)$ and $\tilde c(\mathbf{r},t)$ to satisfy the dynamics of Eq. (2).

To test our theory, we generate all quasi-stationary patterns $\psi(\mathbf{r}), c(\mathbf{r})$ with noise and compute $4kT\mathcal{S}(\psi,c)/\mathcal{T}$ from Eq. (7). In the simulations, parameters similar to those describing experiments on the liquid crystal system 4-n-octylcyanobiphenyl (8CB) [4, 29] are used: $\Gamma_\psi = 1 = \Gamma_c$, $\gamma = 0.73$, $K_\psi = 1.5$, $K_c = 0$ and the pulling speed $v = 0.20$. We take $\Delta T(z) = -T_0$ for $z < -W$, $+T_0$ for $z > W$ and Gz for $|z| < W$ with $G = T_0/W$ [16–18]. The temperature $T_0 = 0.38$ so that $\Delta T(z)$ is greater than the liquidus temperature for $z > W$ and below the solidus for $z < -W$. The simulations are carried out with the width of the temperature variation $W = 50$, the size of the simulation box $L_z = 150$, lattice spacing $\Delta x = 1.0$ and timestep $\Delta t = 0.10$. The system widths are $250 \le L_x \le 1000$ with periodic boundary conditions $\psi(x + L_x, z, t) = \psi(x, z, t)$ and $c(x + L_x, z, t) = c(x, z, t)$ which restricts the allowed values of the $\mathbf{q}$ vector of a periodic variation in the interface between the ordered and disordered phases to be $q_x = n/L_x$ with $n = 1, 2, \cdots L_x$. The Green's function $G(\mathbf{r},\mathbf{r}')$ satisfies $G(x, z; x', z') = G(x + L_x, z; x', z')$ and $G(x, 0; x', z') = 0 = G(x, L_z + 1; x', z')$ and is obtained by numerically inverting $G(\mathbf{p})$, the discrete Fourier transform of $G(\mathbf{r},\mathbf{r}')$. This is used to calculate $\Delta\mathcal{S} \equiv 4kT(\mathcal{S}_n - \mathcal{S}_m)/\mathcal{T}$ where the integers $n, m = q_x L_x$ label the patterns. The results for a system of size $L_x = 500$, $L_z = 150$ are shown in Fig. (1). Note that the action $\Delta S(\psi, c) \propto \eta^2 = \mathcal{O}(T)$. Our $L_x = 500$ system has only three quasi-stationary periodic states in the Eckhaus stable band [26] with $q_x = 0.020, 0.022, 0.024$ corresponding to $n = 10, 11, 12$ cells. The $q_S = 0.020$ minimum action state is conjectured to be the unique selected state. An outstanding question is whether pattern selection also holds in the thermodynamic limit $L_x = \infty$ when the set of stationary states in the Eckhaus stable band is continuous. Comparing with experiment requires extending the calculation to 3D to study the selected stationary structures of the 2D interface [16, 30].

Additional evidence for our conjecture is obtained from simple simulations of Eqs. (1) and (2) from various initial conditions. First, simulations are done in the absence of noise $\eta_\psi = 0 = \eta_c$ with initial values $\psi = \epsilon$ with $-0.50 \le \epsilon \le 0.50$ a uniformly distributed random variable and $c = 0$ for convenience. For system widths $L_x = 250$, 500 and 1000 the system always eventually reaches a periodic stationary state with $q_f = n/L_x = 0.020$. For $L_x = 420$, the final periodicity is $q_f = 8/420 \approx 0.01905$. These results imply that the selected wavevector is the nearest allowed q_f to the preferred value for $L_x \to \infty$. With an initial periodic interface of the form $\zeta(x) = \zeta_0 \cos(2\pi q_0 x)$ with various wavevectors $q_0 = n_0/L_x$ and amplitude ζ_0 similar to that of the final state of the previous simulation, again in the absence of noise. The initial interface is located at the center of the simulation box of size $L_x \times L_z$. Simulations with various allowed values of q_0 and $L_x = 500$ yield final stationary periodic states with $q_x = 0.020, 0.022$ and 0.024 which determine the Eckhaus stable band of wavevectors for our parameter values. In an infinite system, the Eckhaus band has a continuum of q values which is reduced to a small finite number by the finite width $L_x = 500$. When the size is doubled to $L_x = 1000$, we find 7 wavevectors in the Eckhaus stable band from $q_x = 0.019$ to 0.025 as expected.

The next simulations study the effect of noise on the evolution of an initial periodic state with wavevector $q_0 = 0.016$ outside the Eckhaus stable band. Without noise, the evolution is by tip splitting of alternate cells to a final wavevector of $q_f = 0.024$, as shown in Fig. (2). This configuration of 12 cells is different from the expected wavelength of $q_f = 0.020$ as obtained from random initial conditions. With applied noise $\eta_\psi(\mathbf{r}, t)$ independently uniformly distributed with $-\eta < \eta_\psi(\mathbf{r}, t) < +\eta$ with maximum noise amplitude $\eta = 0.10$, the evolution is by a totally different route to the expected state with $q_S = 0.20$ as shown in Fig. (2). This indicates that noise is essential in the selection of the final state and that only one wavenumber in the Eckhaus band is stable with stochastic noise.

Now that we have confirmed the selection of a unique wavelength from almost all initial patterns except for some q_0 in the Eckhaus band, we perform a simulation with an initial patterns $\zeta(x) = \zeta_0 \cos(2\pi q_0 x)$ with $q_0 = 0.024$ with noise to study the stability of modes in the Eckhaus stable band [1, 26]. In the absence of noise, all modes with q_0 in this band are stable. We impose an interface $\zeta(x)$, evolve it in absence of noise for 10^4 time steps to allow higher harmonics to develop and then evolve the system with noise of strength $\eta = 0.20$ for 2×10^5 time steps with results shown in Fig. (3). The interface evolves by the gradual loss of one cell to a final configuration of 11 cells or $q_f = 0.022$ which remains stable for at least 5×10^7 steps. When a brief burst of very strong noise with $\eta > 0.25$ is applied and the system then allowed to evolve under weak noise, the expected $q_S = 0.020$ state results.

These simulation results are consistent with the hypothesis of a unique selected stationary state with a definite periodicity q_S but are not completely conclusive because the $q_0 = 0.022$ state in the Eckhaus stable band does not evolve to the $q_S = 0.020$ within our longest run. If a unique selected state exists, then only one of these should be stable to weak noise but some are stable within our computation time and it is impractical to perform sufficiently long runs to detect an instability toward the hypothetical minimum action selected state with $q_S = 0.020$. This study finds that all states with this single exception do evolve to the minimum action state and, in addition, provides some insight into the relative stability of states in the Eckhaus band. On balance, the evidence for our hypothesis of a unique minimum action selected state seems overwhelming.

One important caveat must be made. In analogy with a finite equilibrium system, we expect all states in our finite system to have *finite* lifetimes as the stochastic noise causes transitions between them. The probability of a particular state (ψ, c) is given by the action of Eq. (7).

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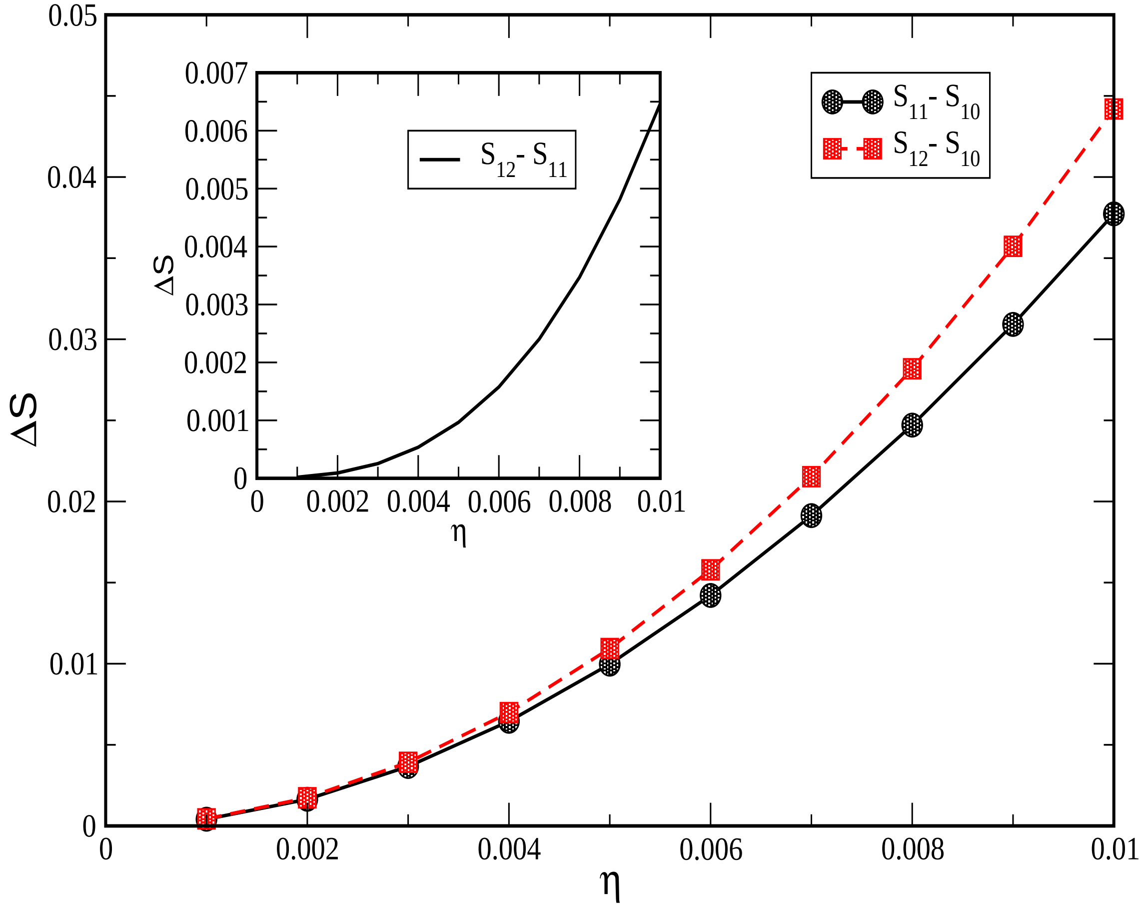

FIG. 1: Action differences $\Delta S = 4kT(S_{11} - S_{10})/\mathcal{T}$ and $4kT(S_{12} - S_{10})/\mathcal{T}$ from Eq. (7) in a system width $L_x = 500$ where the integers $n = q_x L_x$ in S_n. This shows that the pattern with wavelength $q_S = 0.020$ has minimum action.

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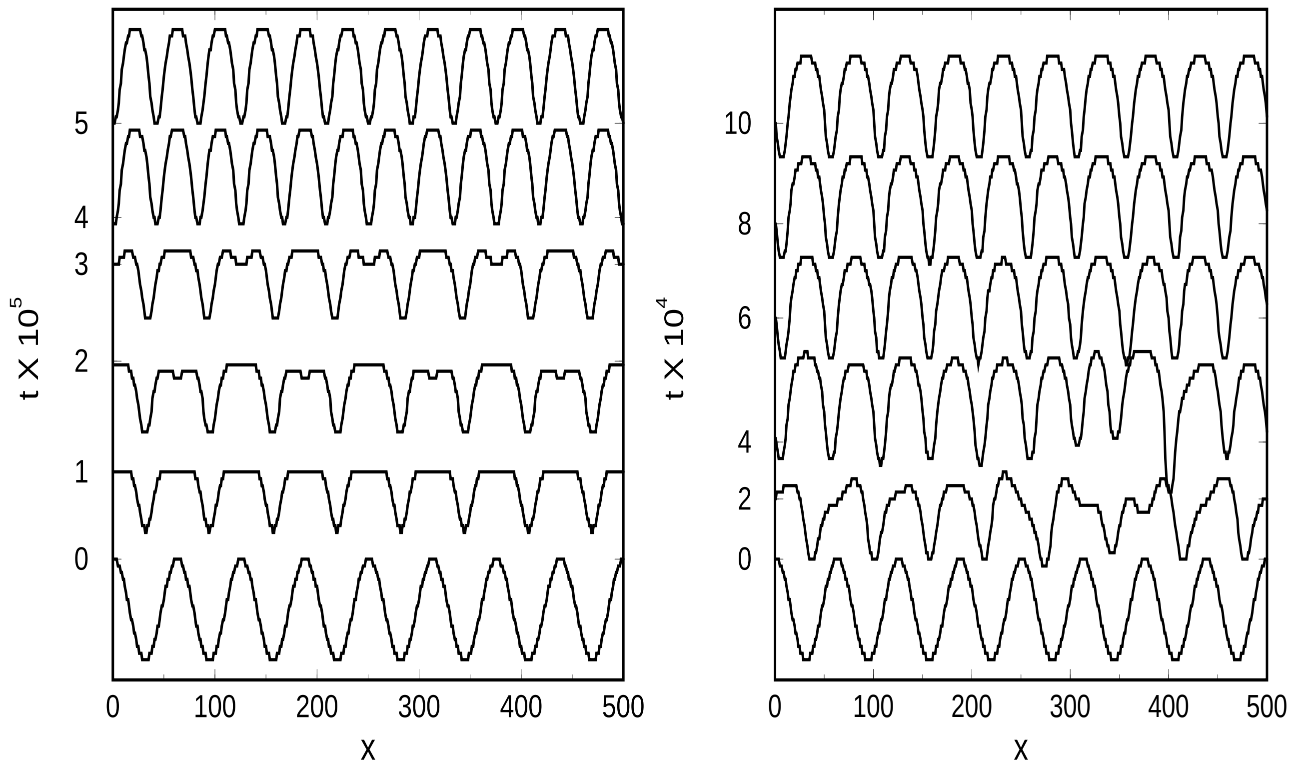

FIG. 2: Left panel: Evolution of cellular pattern of initial periodicity $q_0 = 0.016$ to final state $q_f = 0.024$ with velocity $v = 0.20$ in absence of noise. Right panel: evolution from the same initial configuration in the presence of noise of strength $\eta = 0.10$ to a final state with $q_f = 0.020$. In both cases, $L_x = 500$.

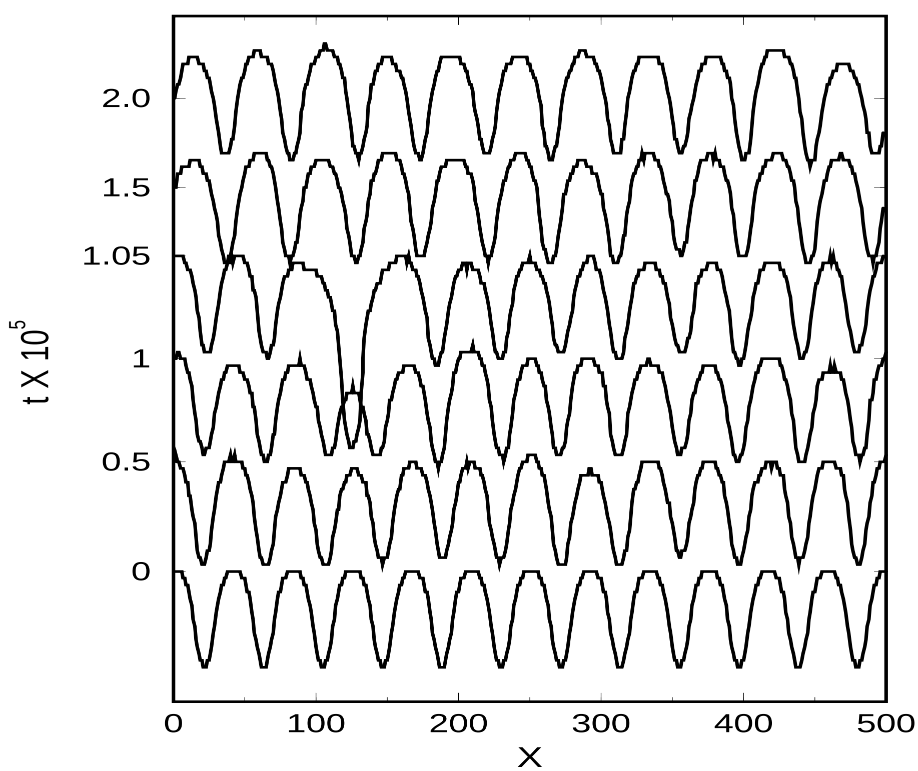

FIG. 3: Evolution of interface with $q_0 = 0.024$ with noise of strength $\eta = 0.20$ to final state $q_f = 0.022$. $L_x = 500$.