Self-Assembly and Convergence Rates of Heterogeneous Reversible Growth Processes (Extended Abstract)

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1 Introduction

In tile-based self-assembly models, tiles are constructed so that specified pairs are encouraged or discouraged to join together. This model was first introduced by Wang [11] and was shown to be a universal model of computation. Winfree [12] and others studied the model in the context of DNA computation by constructing tiles made of double-crossover strands, where the sides of each tile could be encoded with single-stranded DNA and tiles are more or less likely to form bonds according to the number of complementary base pairs on their corresponding sides. See, e.g., [5, **??**, 12] for more details. Here we are concerned with determining the rate of convergence to equilibrium of some simple self-assembly processes.

In particular, we consider a lattice-based growth model consisting of three types of tiles: *a seed tile* placed at the origin, *border tiles* that form the left and bottom border of an $n \times n$ region, and *interior tiles* that can be placed in the remaining part of the region. We assume that the border forms quickly after a seed tile appears, and we restrict to a growth model in which interior tiles can attach to the aggregate if their left and bottom neighbors are present, whereas they can detach if their right and upper neighbors are absent. It is easy to see that the only shapes that can be formed are tiles that are neatly packed into the lower-left corner of the region so that the upper border forms a *staircase*

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walk that travels to the right and down (see Fig. 1). We refer to the ratio of the attach and detach rates as the *bias* of the growth process.

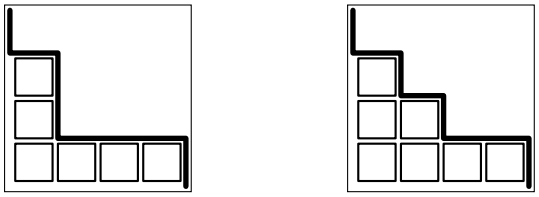

Fig. 1. A pair of tilings connected by a single move of the Markov chain.

Variants of this growth model have been previously considered in order to study various aspects of self-assembly. For example, Baryshnikov et al. [2] consider a non-reversible version of this process when n is large to determine the limiting shape, and note that the behavior is captured by a TASEP (totally asymmetric simple exclusion process). Majumder et al. [8] initiated the study of the reversible process in the finite setting where tiles are allowed to detach as well as attach to the aggregate.

It has been noted for many models that dynamics of self-assembly can be viewed as a Markov chain, and the convergence rate of the chain captures the rate that the assembly process tends towards equilibrium (see, e.g., [1, 7, 8, 6]). Majumder et al. [8] prove that the reversible growth process described above is rapidly mixing (i.e., it converges in time polynomial in n) in two and three dimensions if tiles attach at a much faster rate than they detach. In fact, the same two-dimensional process had been studied previously by Benjamini et al. [3] in the context of biased card shuffling and biased exclusion processes. They give optimal bounds on the mixing time of the chain of $O(n^2)$ for all fixed values of the bias. Subsequently, Greenberg et al. [7] discovered an alternative proof establishing the optimal bound on the mixing rate for any fixed bias in two dimensions. This new proof is much simpler, and moreover it can be generalized to higher dimensional growth processes provided the bias is not too close to one.

A common feature of all of these previous results is that all interior tiles are always treated identically. Thus, the rate at which a tile is allowed to attach or detach from the large aggregate is independent of the position of the tile. This is a severe shortcoming since in reality several properties of the growth processes can affect these rates, including the particular encodings on the sides of the double-crossover molecules comprising the tiles, as well as the relative densities of different tiles that can be attached in various positions. Mathematically, the problem becomes more intriguing as well. Physicists refer to this property whereby the bias is location dependent as "fluctuating bias," and it has been noted that methods that allow us to analyze systems with fixed bias do not readily generalize to the fluctuating setting.

In this paper we consider the heterogeneous setting where the bias of a tile depends on its location. The state space Ω us the set of valid tilings of the $n \times n$ region where the upper/right boundary is a staircase walk starting at $(0, n)$ and ending at $(n, 0)$. Let $\alpha(x, y)$ and $\beta(x, y)$ be the probabilities of adding and removing a tile at position (x, y) for the heterogeneous model. We call $\lambda_{x,y} = \alpha(x, y)/\beta(x, y)$ the *bias* at position (x, y).

The self-assembly Markov chain $\mathcal{M}_{SA}$ tries to add or remove individual tiles in each step and is formalized below. We give the first rigorous proofs that the chain is rapidly mixing for various settings of the $\{\lambda_{x,y}\}$. In particular, we derive optimal bounds on the mixing time if $\lambda_{x,y} > 4$ for all (x, y) and we give polynomial bounds on the mixing time when $\lambda_{x,y} \geq 2$.

2 The Mixing Time of the Self-Assembly Markov Chain

The growth process is defined so that the rate of adding a tile at position (x, y) is $\alpha(x, y)$ and the rate of removing that tile is $\beta(x, y)$. We are interested in the case where $\lambda_{x,y} = \alpha(x, y)/\beta(x, y) \geq 1$ for all x, y. Since the chain is reversible, there is a unique stationary distribution, and it is straightforward to see the chain converges to

$$\pi(\sigma) = \frac{\prod_{(x,y)\in\sigma} \lambda_{x,y}}{Z},$$

where $Z = \sum_{\tau\in\Omega} \prod_{(x,y)\in\tau} \lambda_{x,y}$ is the normalizing constant. Our goal is to sample configurations in Ω according to the distribution π.

We can now formalize the Markov chain $\mathcal{M}_{SA}$. At each time step, we pick a vertex v uniformly along the upper border of the aggregate, each with probability $1/(2n)$ since the border has length $2n$. It will be convenient in the analysis to add a self-loop probability at each position, so we flip a coin and if the coin lands on heads we try to add a tile at v and if the coin is tales we try to remove the tile at v.

To define the transition probabilities of $\mathcal{M}_{SA}$, we use the Metropolis-Hastings probabilities [9] with respect to π. The transitions are

$$\mathcal{M}_{SA}(\sigma, \tau) = \begin{cases} 1/(4n), & \text{if } \tau = \sigma \cup t_{x,y} \text{ for some tile } t_{x,y}; \\ 1/(4n\lambda_{x,y}), & \text{if } \sigma = \tau \cup t_{x,y} \text{ for some tile } t_{x,y}; \\ 1 - \sum_{\tau'\neq\tau} \mathcal{M}_{SA}(\sigma, \tau'), & \text{if } \sigma = \tau; \\ 0, & \text{otherwise.} \end{cases}$$

The time a Markov chain takes to converge to its stationary distribution, or the *mixing time*, is measured in terms of the total variation distance between the distribution at time t and the stationary distribution (see, e.g., [7]). Our goal is to show that the mixing time of the chain $\mathcal{M}_{SA}$ is bounded by a polynomial in the size of the region being tiled. We can show this provided the lower bound on the bias is not too close to one. We briefly state our results and outline the two general techniques used, deferring complete proofs for the full version of the paper. We believe these approaches can be generalized to many other self-assembly models.

2.1 Coupling Using A Geometric Distance Function

Theorem 1 *Let $\lambda_L \geq 1$ be a lower bound on the bias and let λ_U be an upper bound satisfying*

$$\lambda_U := \begin{cases} (\frac{2}{\sqrt{\lambda_L}} - 1)^{-1} & \text{if } \lambda_L < 4 \\ \infty & \text{if } \lambda_L \geq 4 \end{cases}.$$

If, for all x, y, we have $\lambda_L < \lambda_{x,y} < \lambda_U$, then the mixing time of $\mathcal{M}_{SA}$ satisfies $\tau(\epsilon) = O(n^2 \ln \epsilon^{-1})$.

In particular, this theorem gives the optimal bound on the mixing time when the lower bound λ_L on the bias everywhere is at least four.

This result is obtained by the path coupling method (see [4]) using a carefully defined distance metric. As in [7] which deals with the homogeneous case (i.e., fixed λ), we use a geometric distance metric, with the following additional definition. For any tile $\overline{v} = (x, y)$, let $\mathrm{diag}(\overline{v}) = x + y$. Then we define the distance function ϕ to be

$$\phi(\sigma, \rho) = \sum_{\overline{v}=(x,y)\in\sigma\oplus\rho} \gamma^{\mathrm{diag}(\overline{v})-2n},$$

where $\gamma = \frac{1}{2}(1 + \frac{1}{\lambda_U})$. We can now verify that any pair of tilings at Hamming distance 1 (e.g., Fig 2 (a)) ϕ is decreasing in expectation during moves of the coupled chain. This is sufficient to bound the mixing time of the chain following the arguments in [7].

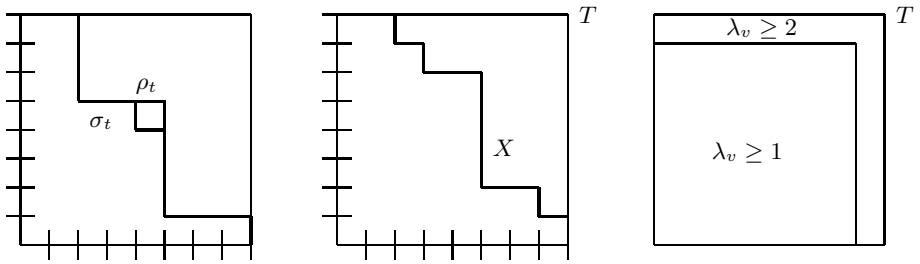

Fig. 2. (a) Example of tilings at Hamming distance 1; (b) A staircase walk with 5 good (increasing) moves and 4 bad (decreasing) moves with respect to the highest tiling T; (c) A region R where the tiles along the upper boundary have bias greater than 2, whereas the other tiles are unrestricted.

2.2 Hitting Time to the Maximal Tiling

Using a different approach based on hitting times we can show that the chain is rapidly mixing provided the bias everywhere is at least 2.

Theorem 2 *Suppose that for all positions (x, y), $2 \leq \lambda_L$. Then the mixing time of $\mathcal{M}_{SA}$ satisfies*

$$\tau(\epsilon) = O(n^3 \ln(n\epsilon^{-1})).$$

The proof of this theorem relies on the monotonicity of $\mathcal{M}_{SA}$ with respect to the trivial coupling. In other words, if (σ_t, ρ_t) are coupled and $\sigma_t \subseteq \rho_t$, then after one step of the coupling, $\sigma_{t+1} \subseteq \rho_{t+1}$. This implies that the coupling time is bounded by the time to hit the highest configuration T starting from the bottom configuration B, and we can show that this will happen quickly because the distance to T is always non-increasing in expectation (see Fig. 2 (b)).

In fact, something more general is true. The tile biases can vary widely everywhere, as long as the upper boundary of the region has bias at least $\lambda_L = 2$ (see Fig. 2 (c)). This surprising result says that the pull of the upper boundary tiles is strong enough to ensure a fast hitting time to the highest configuration.

Theorem 3 *Let R be an $n \times n$ region. Suppose that for every $(x, y) \in R$ the bias $\lambda_{x,y} > 1$. Define the upper border $S = \{(x, n) \in R\} \cup \{(n, y) \in R\}$ and suppose that for all $(x, y) \in S$, we have $\lambda_{x,y} > 2$. Then*

$$\tau(\epsilon) = O(n^3 \ln(n\epsilon^{-1})).$$

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