Structural DNA nanotechnology

a.k.a. DNA carpentry

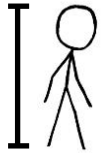
a.k.a. DNA self-assembly

ECS 232: Theory of Molecular Computation, UC Davis

Ljubljana Marshes Wheel. 5k years old

Building things

Newgrange, Ireland. 5.2k years old

Building things by hand: use tools! Great for scale of $10^{\pm 2} \times$ 

[slides credit: Damien Woods]

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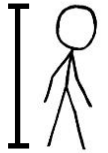
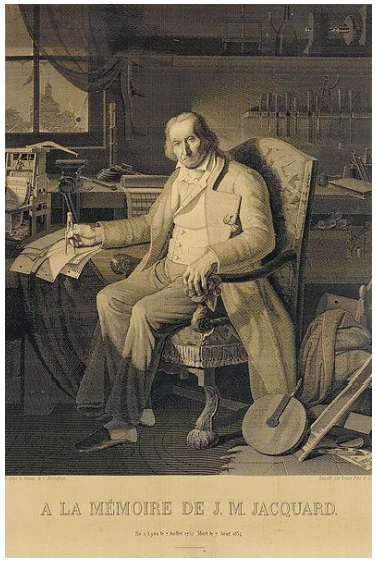
Building things by hand: use tools! Great for scale of $10^{\pm 2} \times$ 

Building tools that build things: specify target object with a computer program



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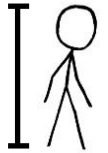
Building things by hand: use tools! Great for scale of $10^{\pm 2} \times$ 

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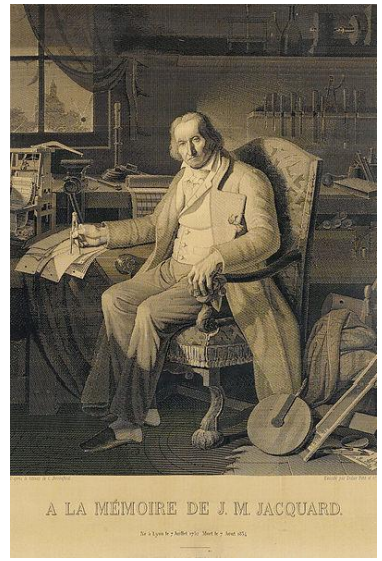
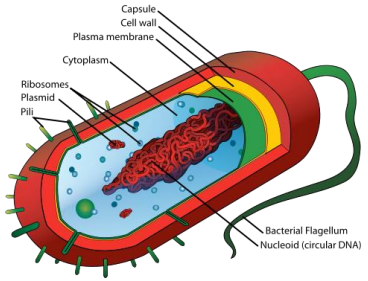


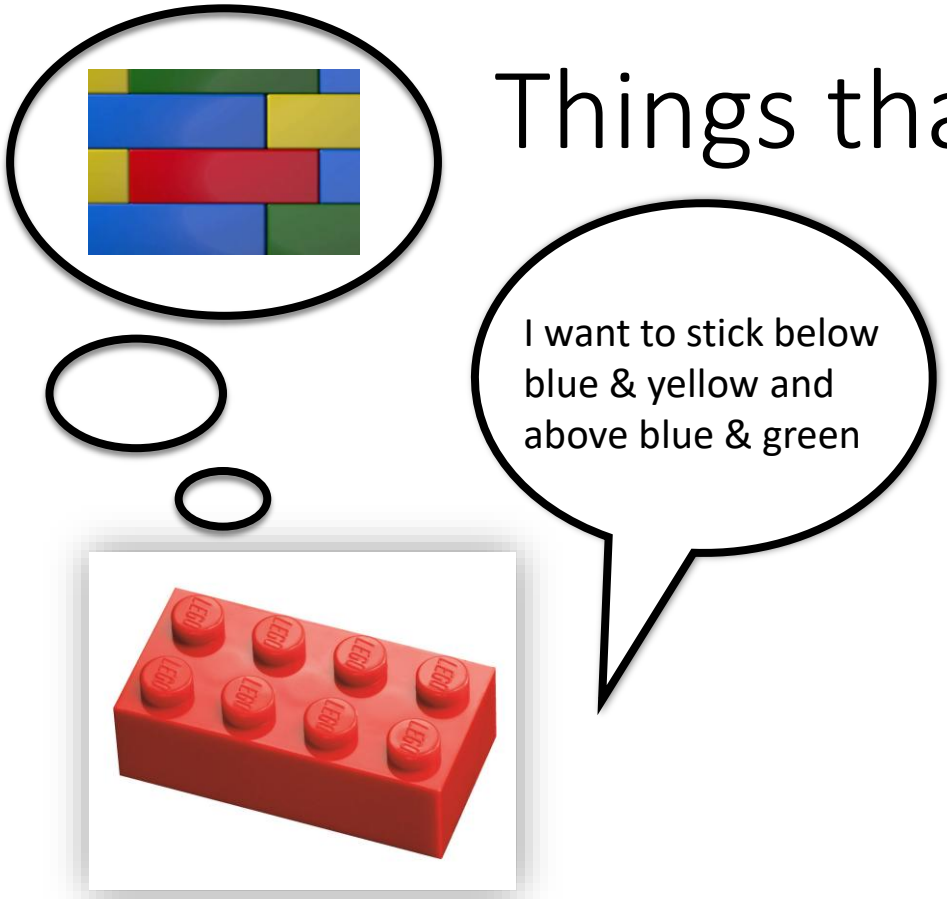


Mariana Ruiz Villarreal

Programming **things to build themselves**: for building in small wet places where our hands or tools can't reach

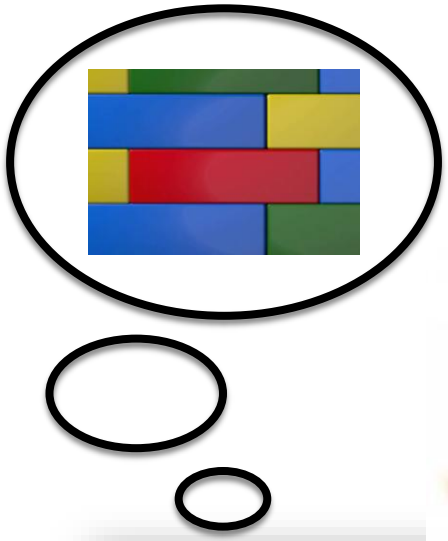
[slides credit: Damien Woods]

Things that build themselves



Our topic: self-assembling molecules that compute as they build themselves

[slides credit: Damien Woods]



Things that build themselves

Our topic: self-assembling molecules that compute as they build themselves

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Things that build themselves

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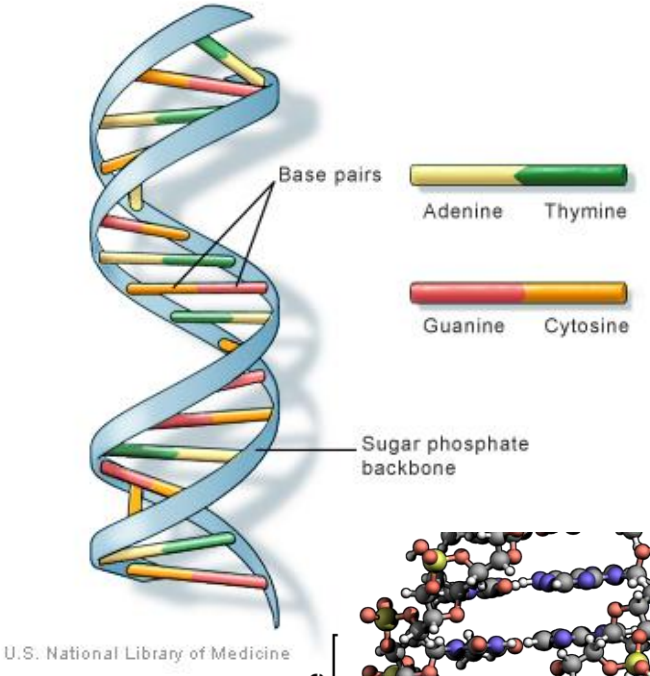
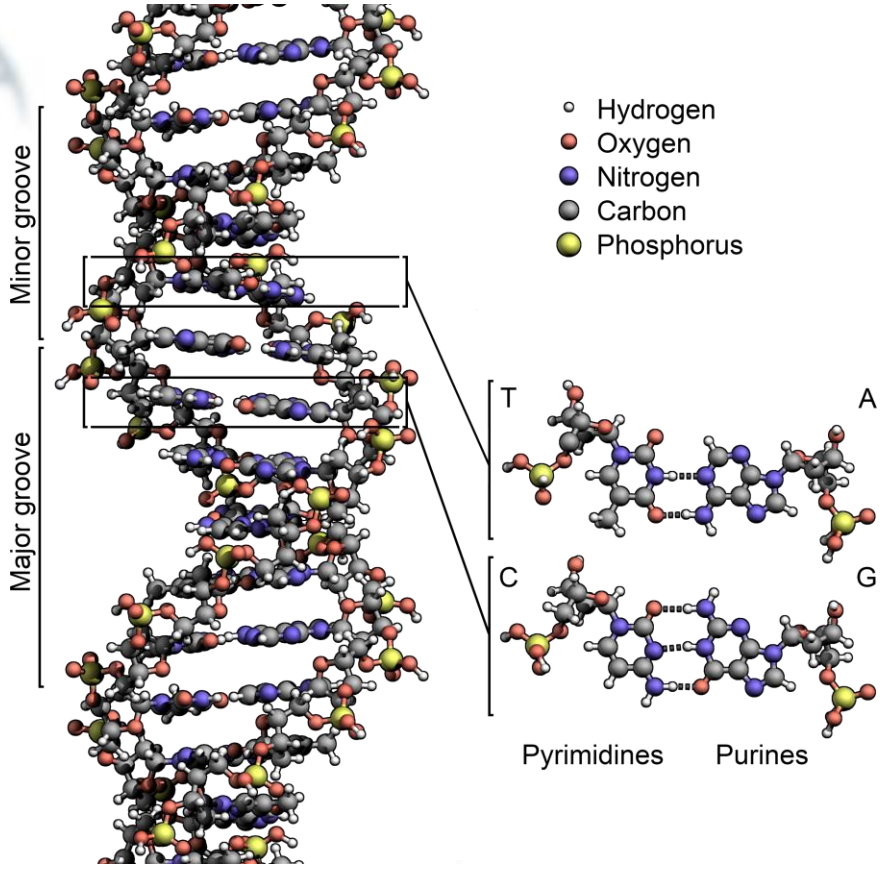
DNA as a building material





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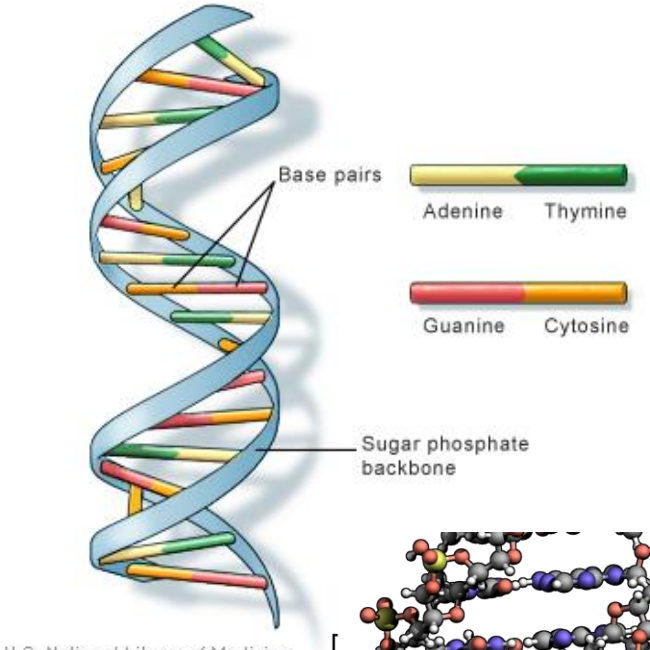

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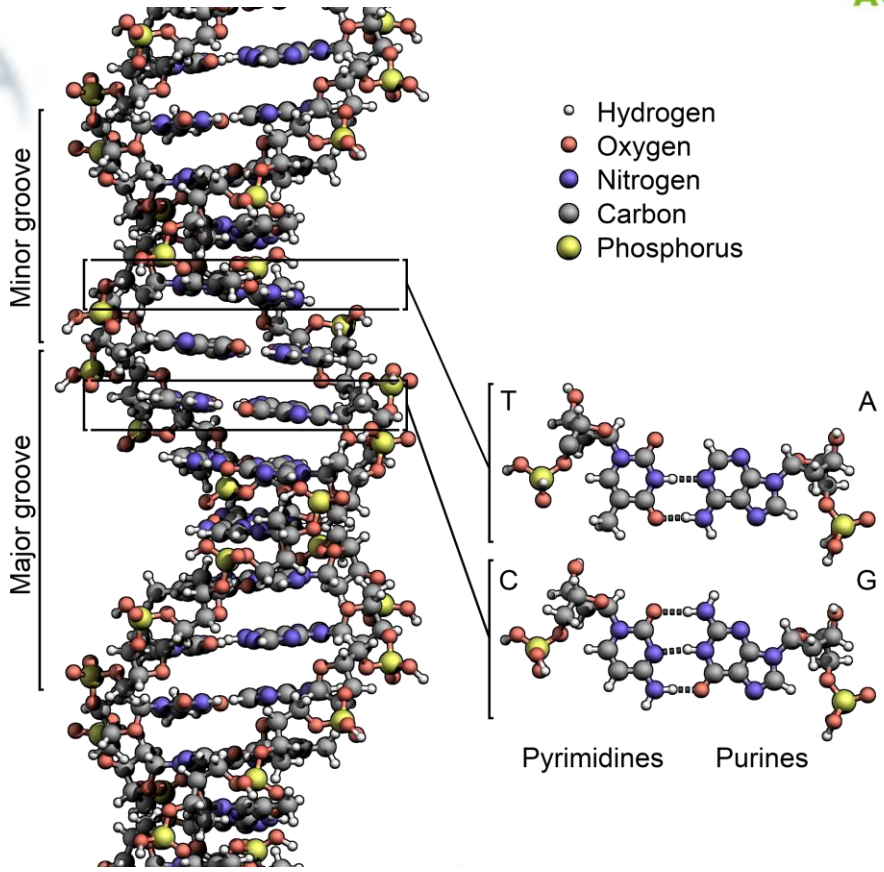
TCGGAAATAAAATCGGAC

AGCCTTTATTTTAGCCTG



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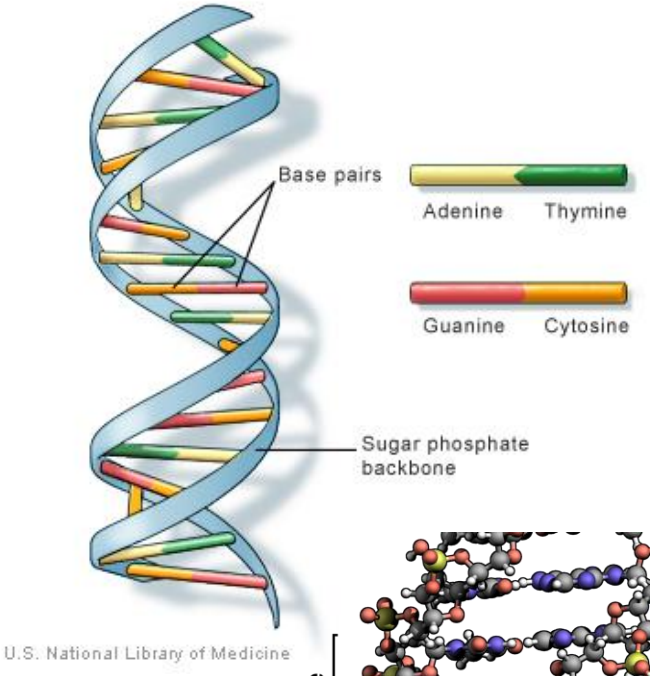

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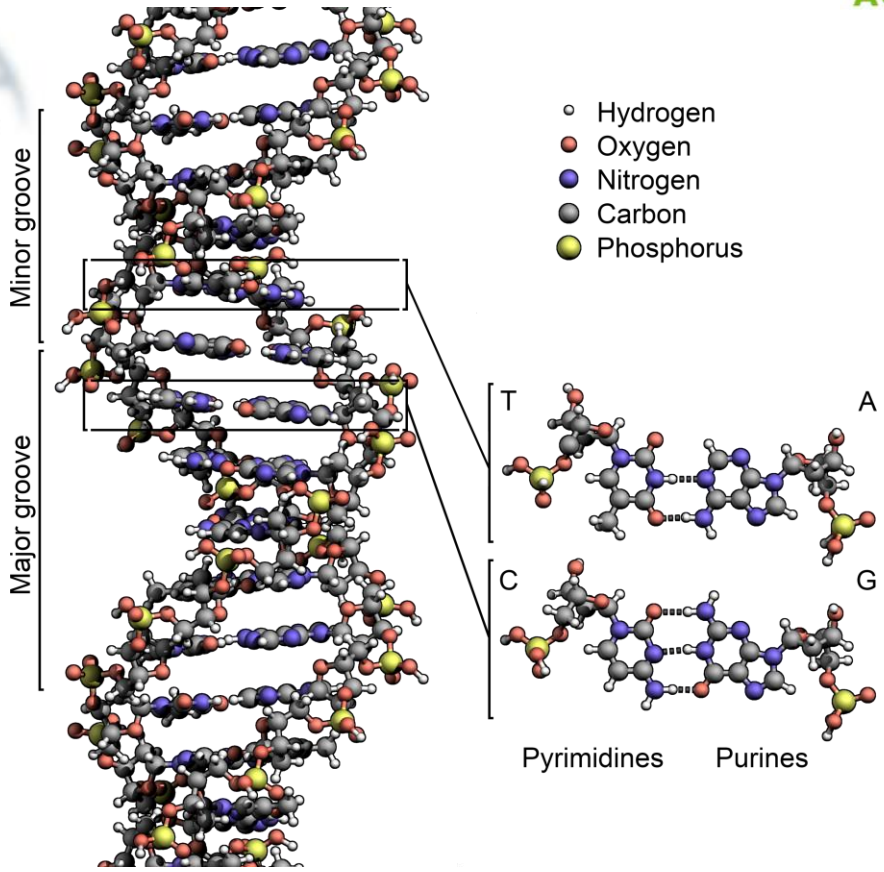
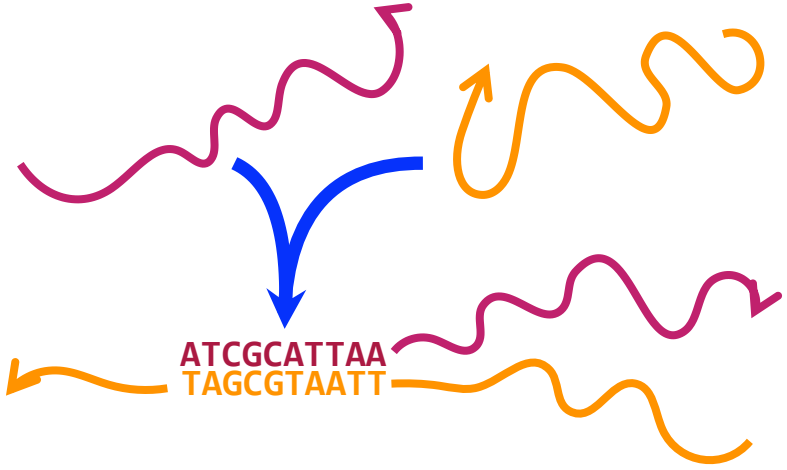


DNA strands bind even if only *part* of strands are complementary:



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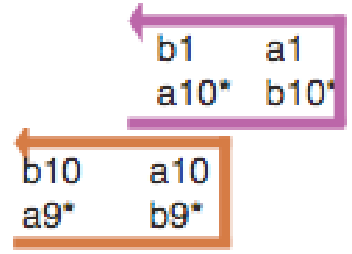
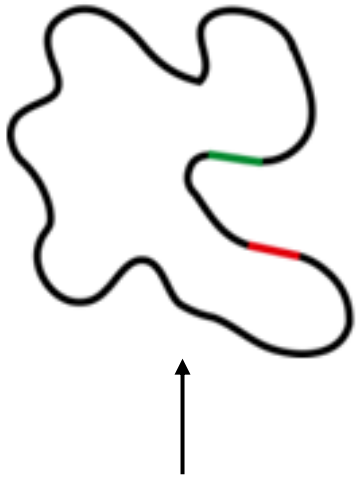


DNA origami

Paul Rothemund
Folding DNA to create nanoscale shapes and patterns
Nature 2006

DNA origami



scaffold DNA strand

(M13mp18 bacteriophage virus)

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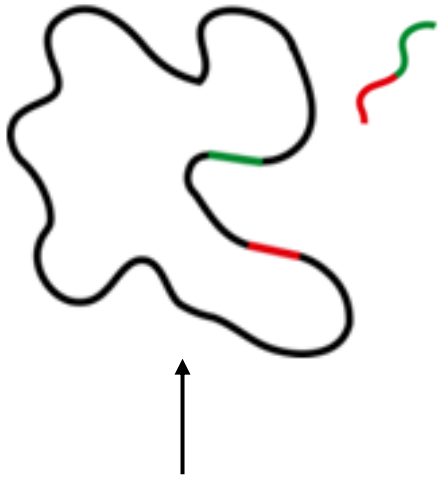

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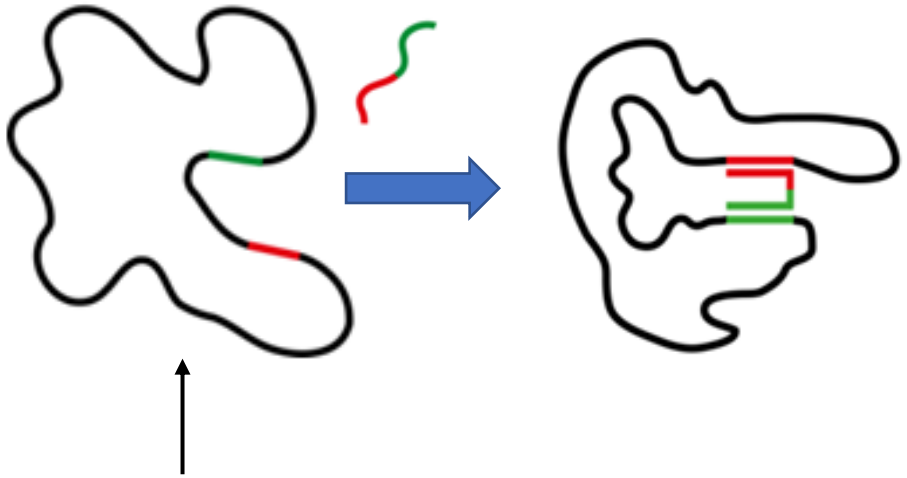

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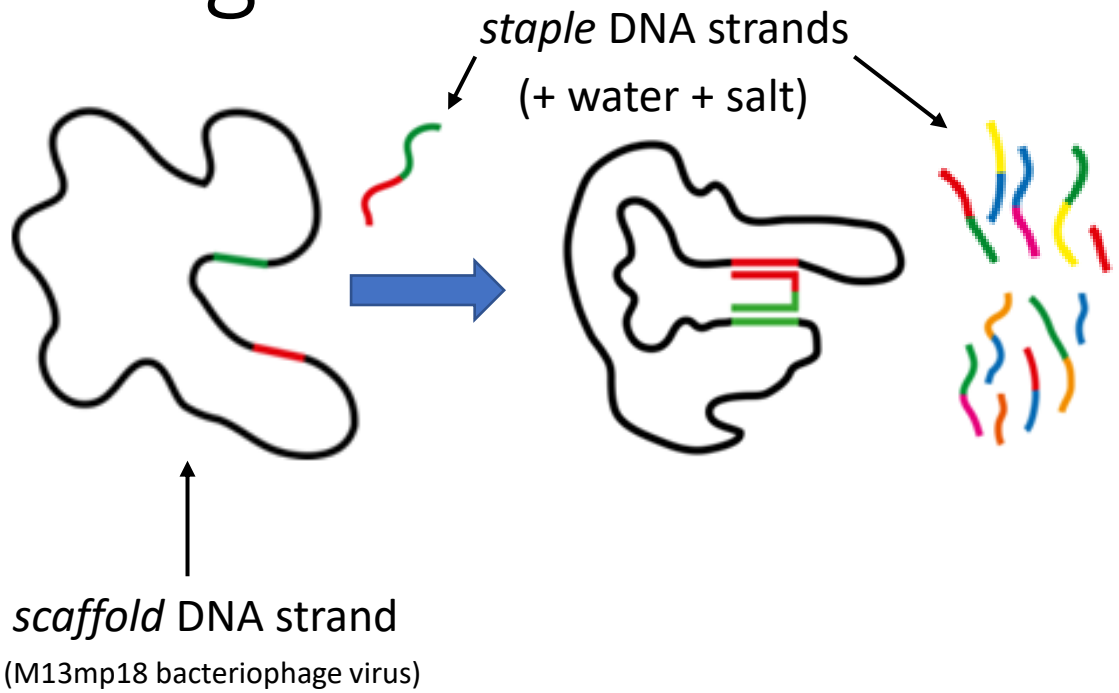

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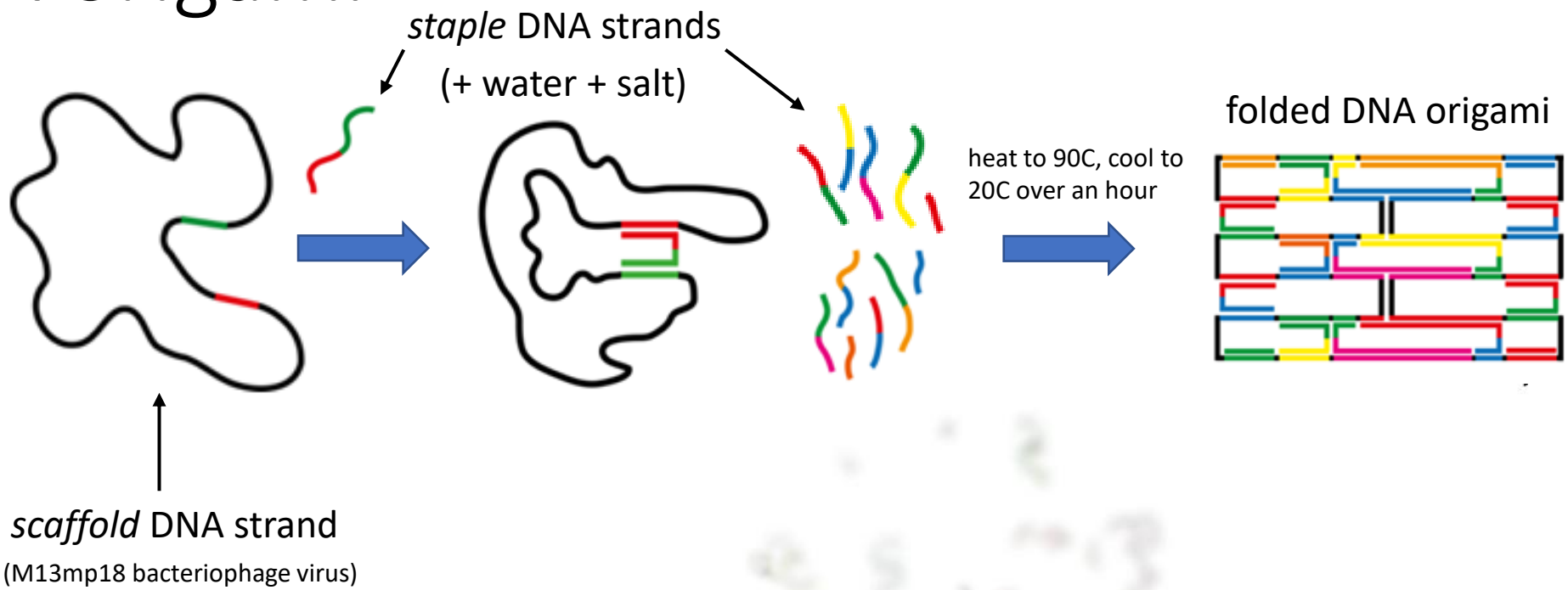

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© http://openwetware.org/wiki/Biomod/2014/Design

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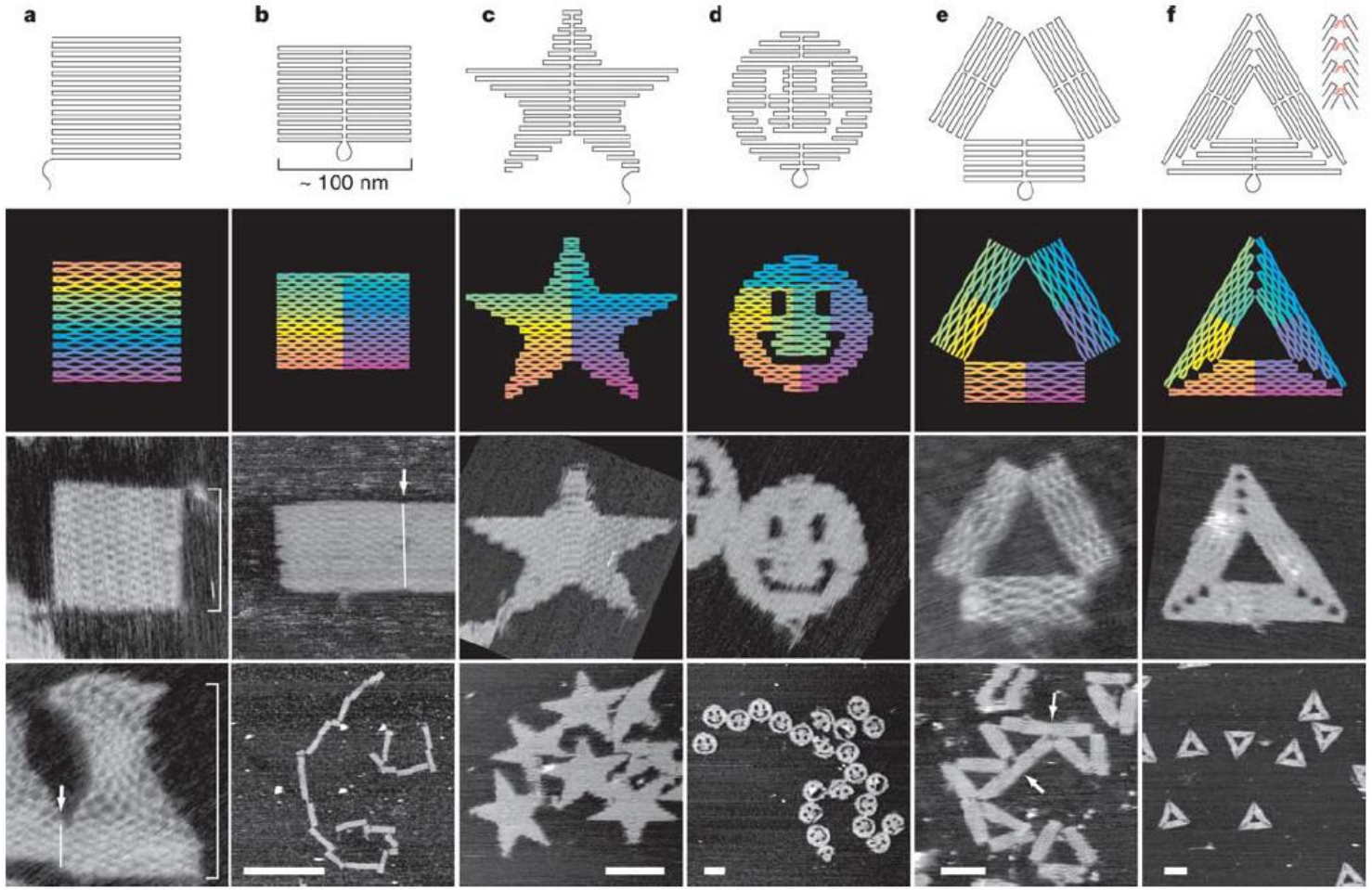
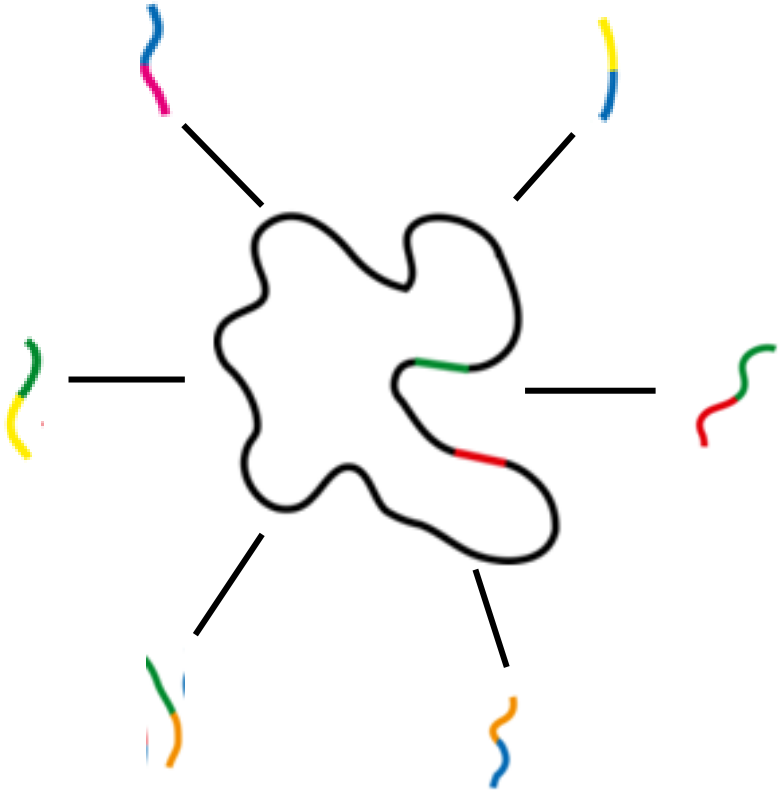


Atomic force microscope images

100 nm

Binding graphs

DNA origami: **star graph**
(all binding is between staples and scaffold)



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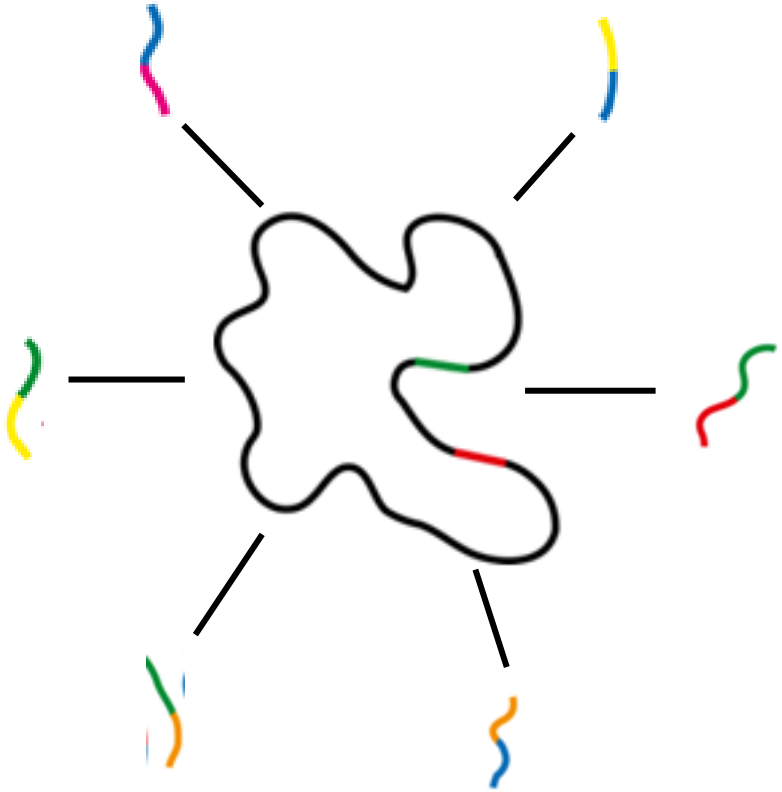
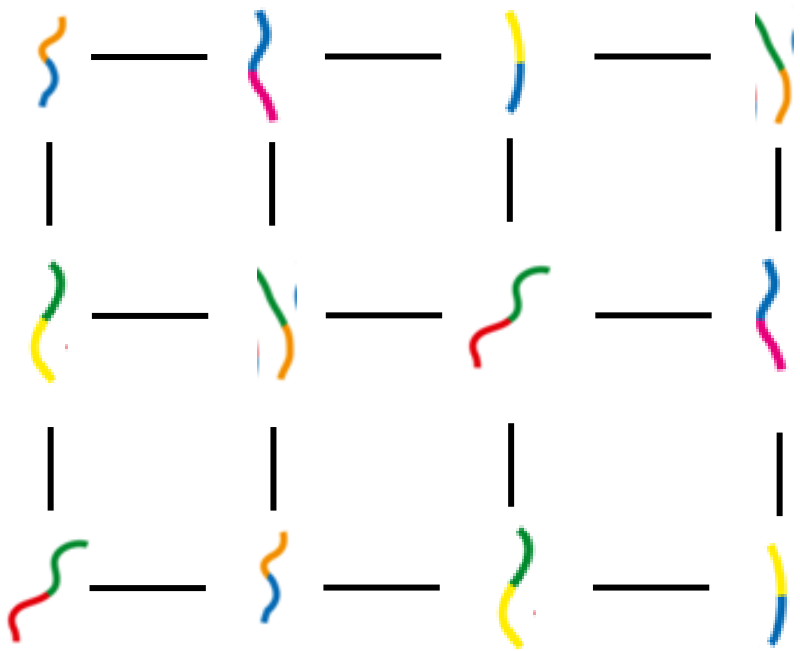


DNA tiles: **grid graph**
(tiles bind to each other, each has ≤ 4 neighbors)

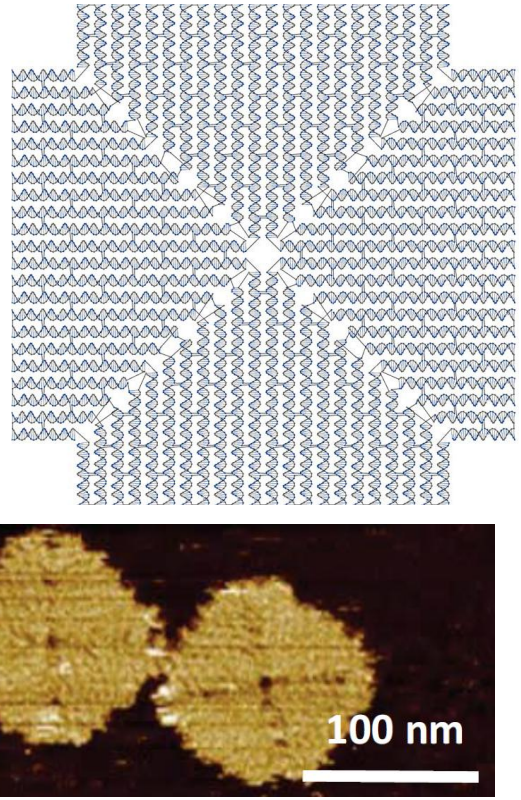


DNA tile self-assembly

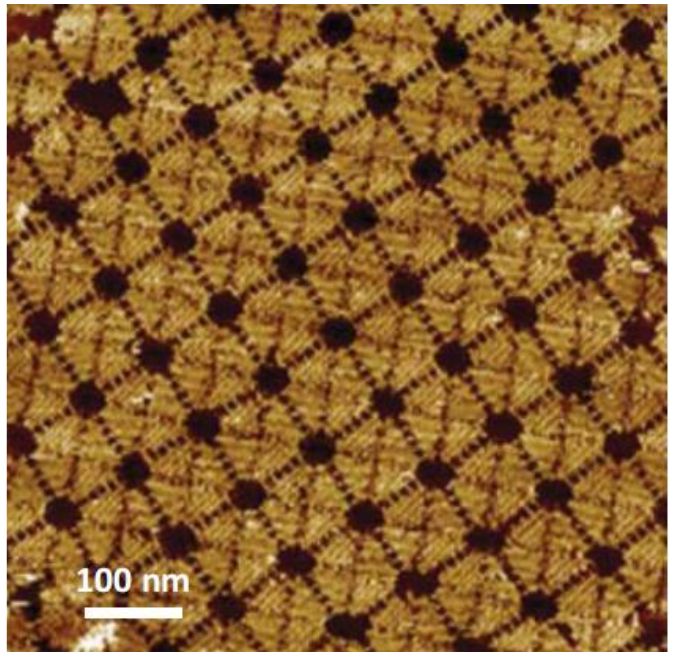
DNA tile self-assembly

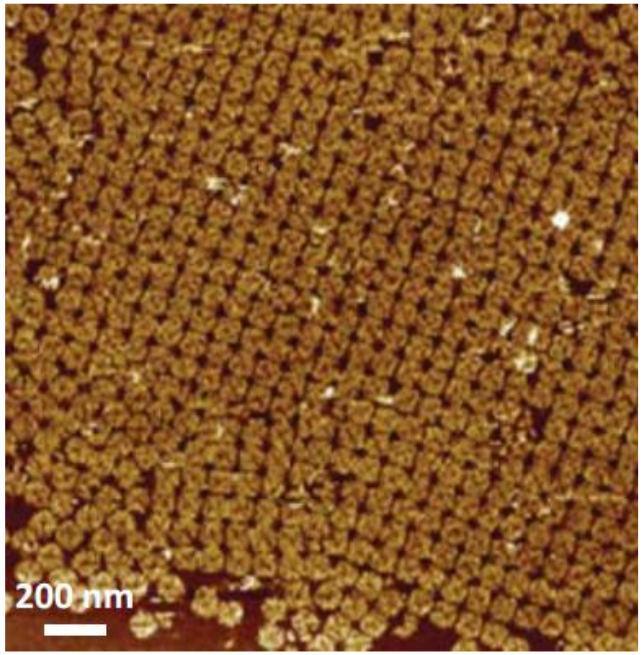
monomers (“tiles” made from DNA) bind into a crystal lattice

tile



lattice

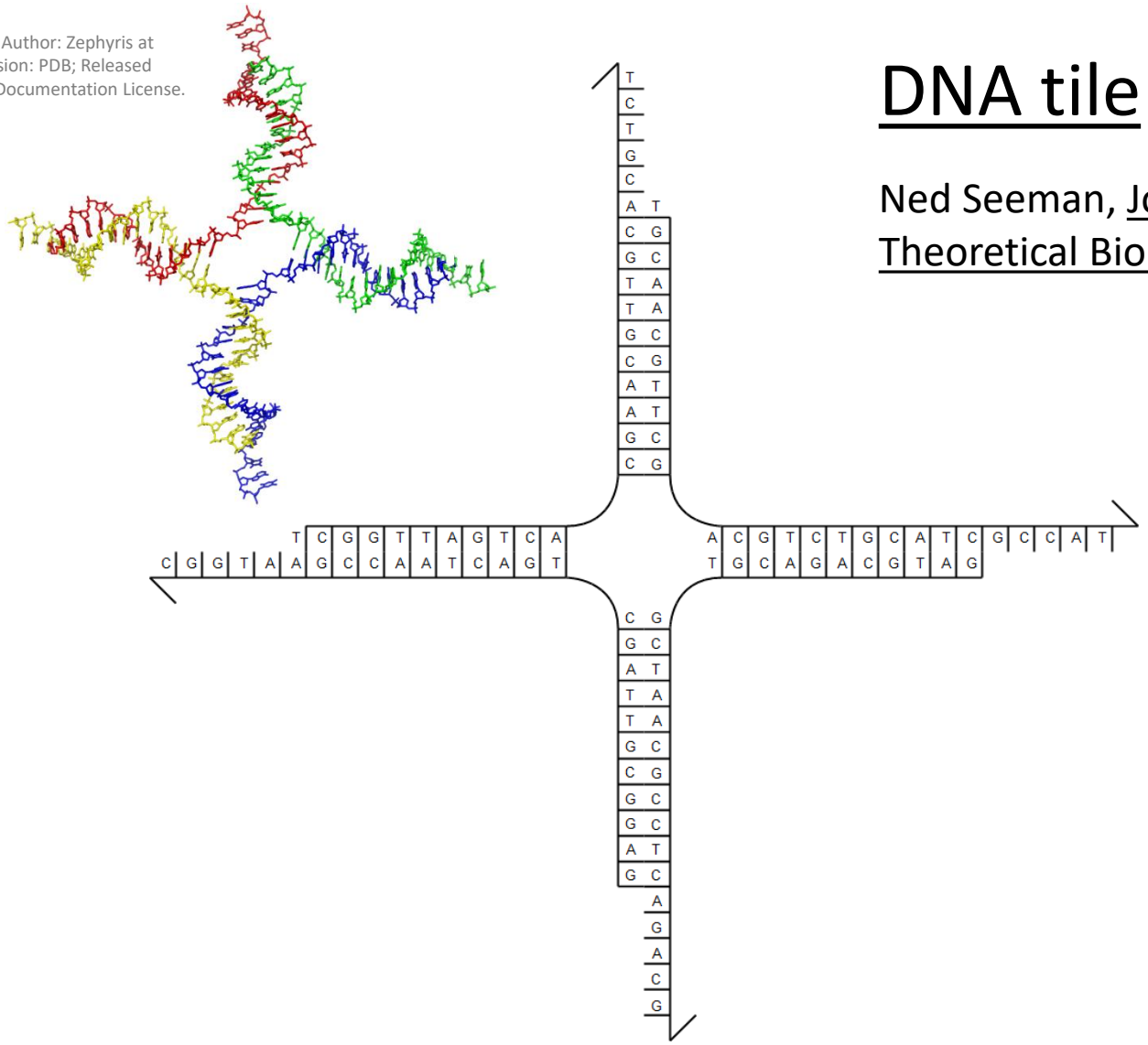




Source: *Programmable disorder in random DNA tilings*. Tikhomirov, Petersen, Qian, Nature Nanotechnology 2017

Practice of DNA tile self-assembly

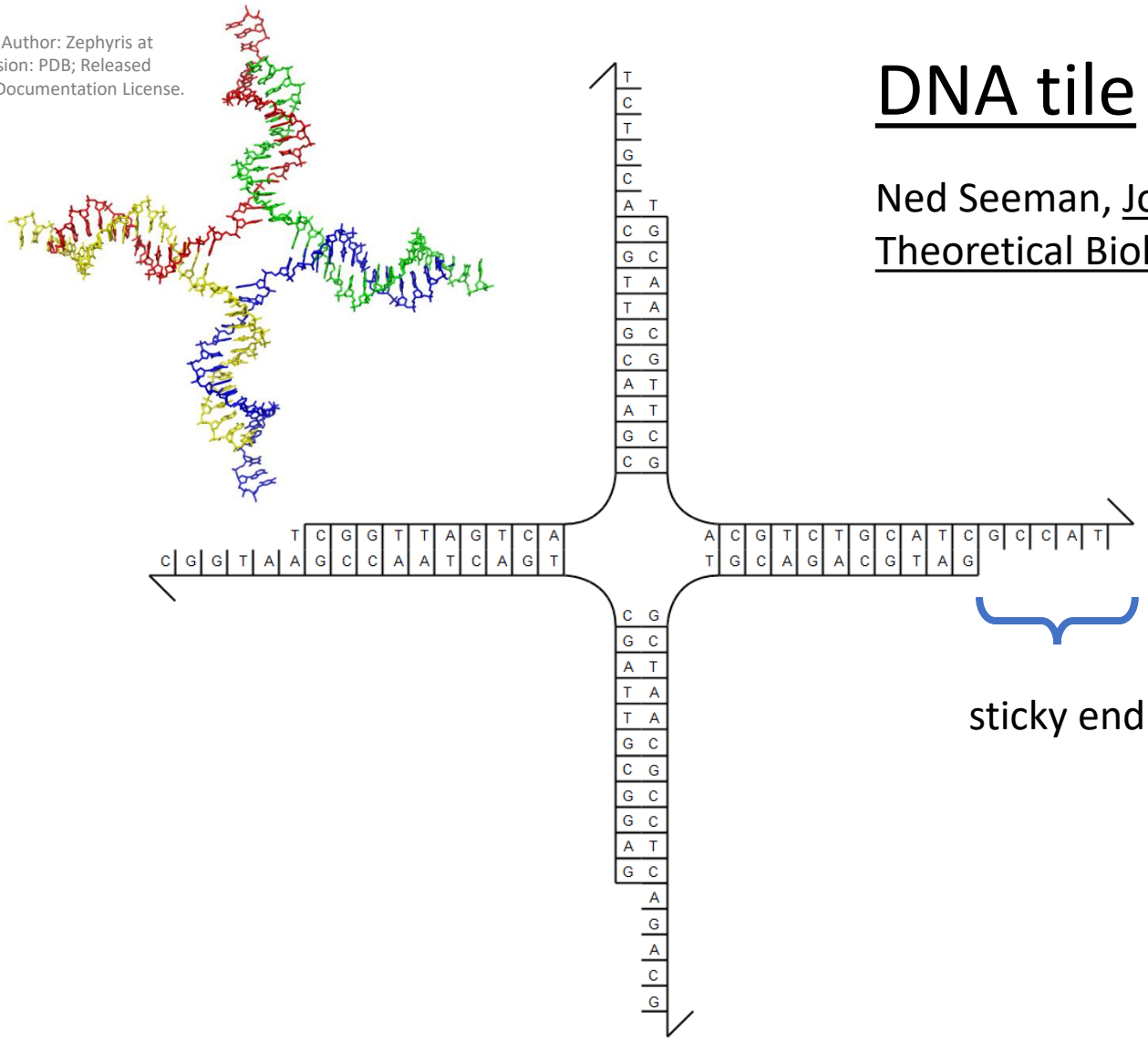
Source:en.wikipedia; Author: Zephyris at en.wikipedia; Permission: PDB; Released under the GNU Free Documentation License.



DNA tile

Ned Seeman, Journal of Theoretical Biology 1982

Practice of DNA tile self-assembly



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sticky end

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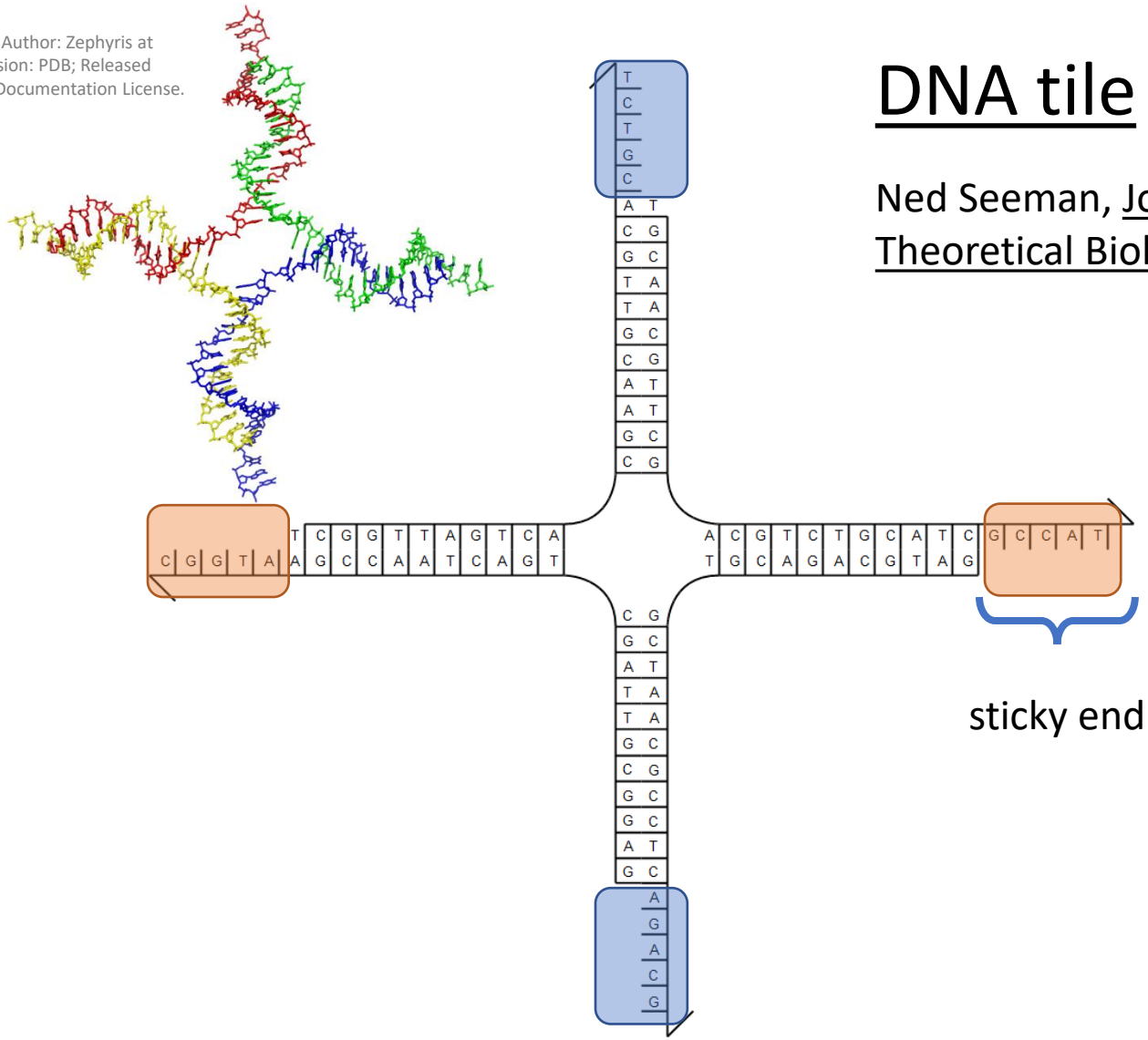


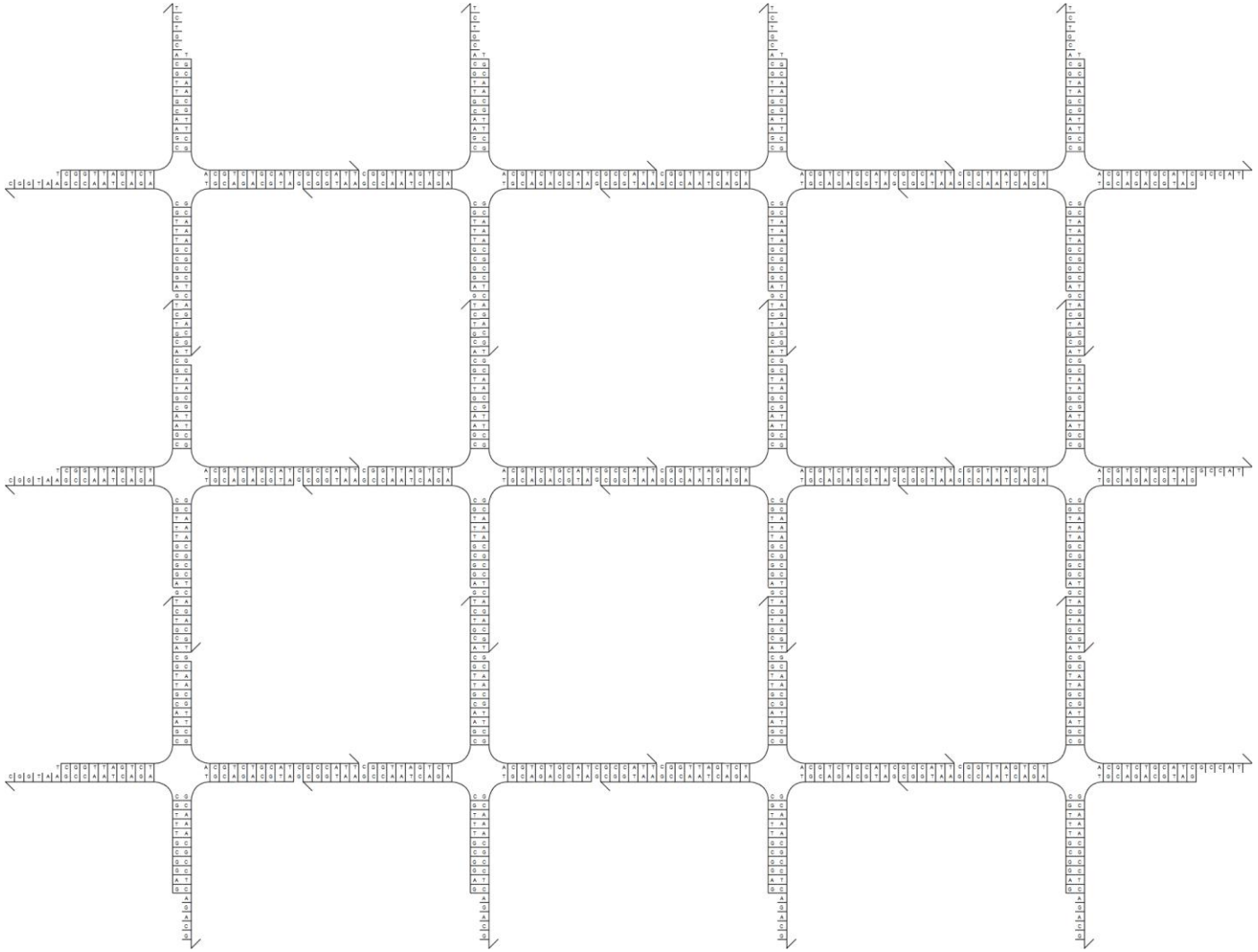
DNA tile

Ned Seeman, Journal of Theoretical Biology 1982

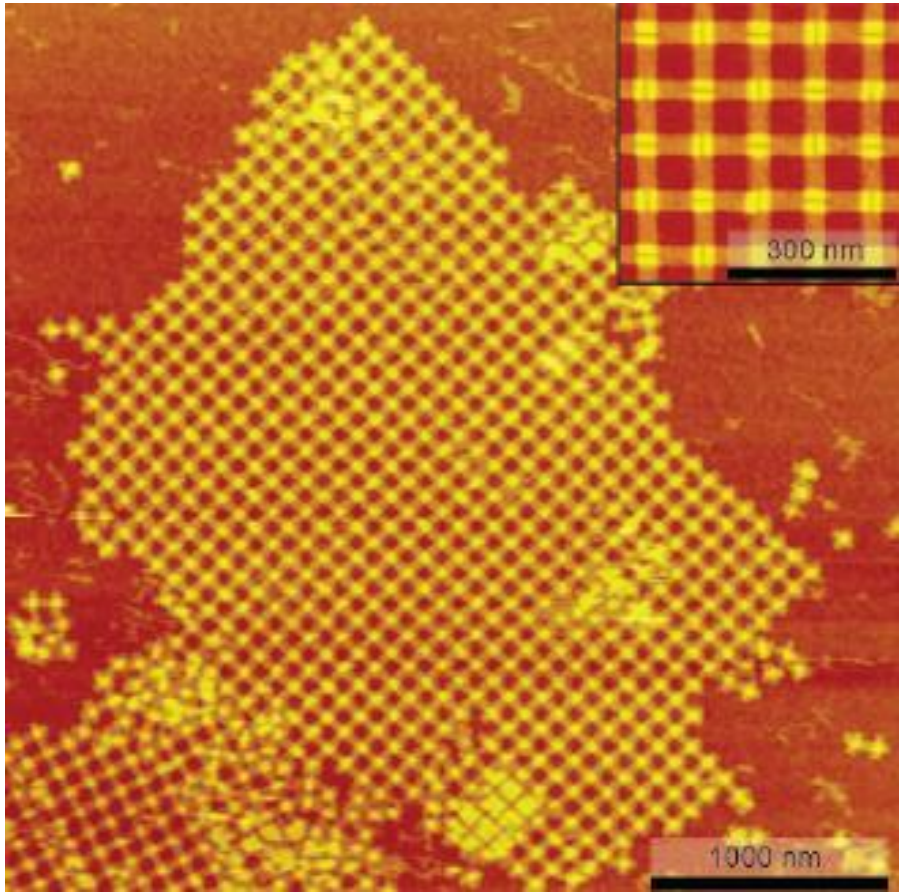
sticky end

Practice of DNA tile self-assembly

Place many copies of DNA tile in solution…



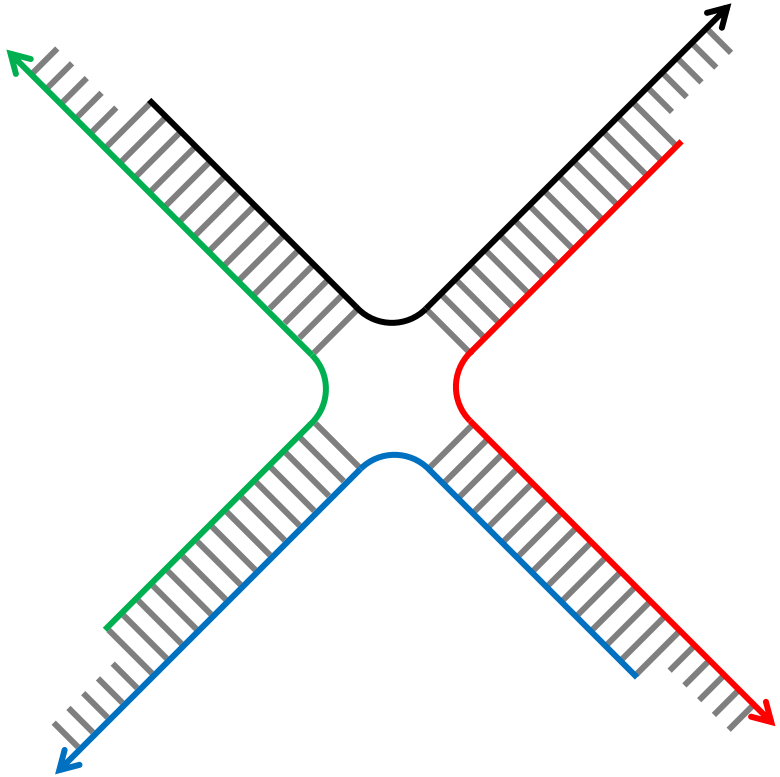
(not the same tile motif in this image)



Liu, Zhong, Wang, Seeman, Angewandte Chemie 2011

Practice of DNA tile self-assembly

What really happens in practice to Holliday junction (“base stacking”)



Practice of DNA tile self-assembly

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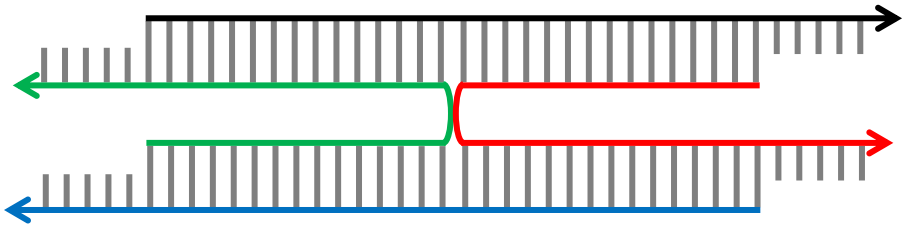

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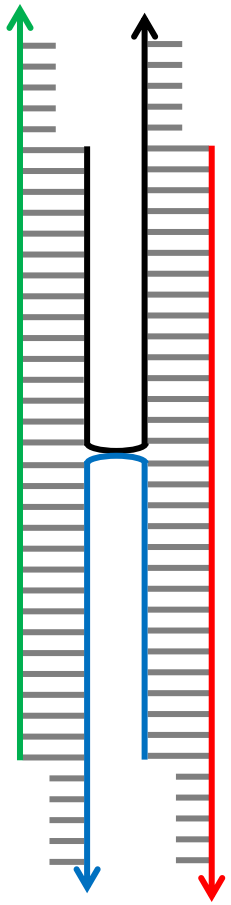

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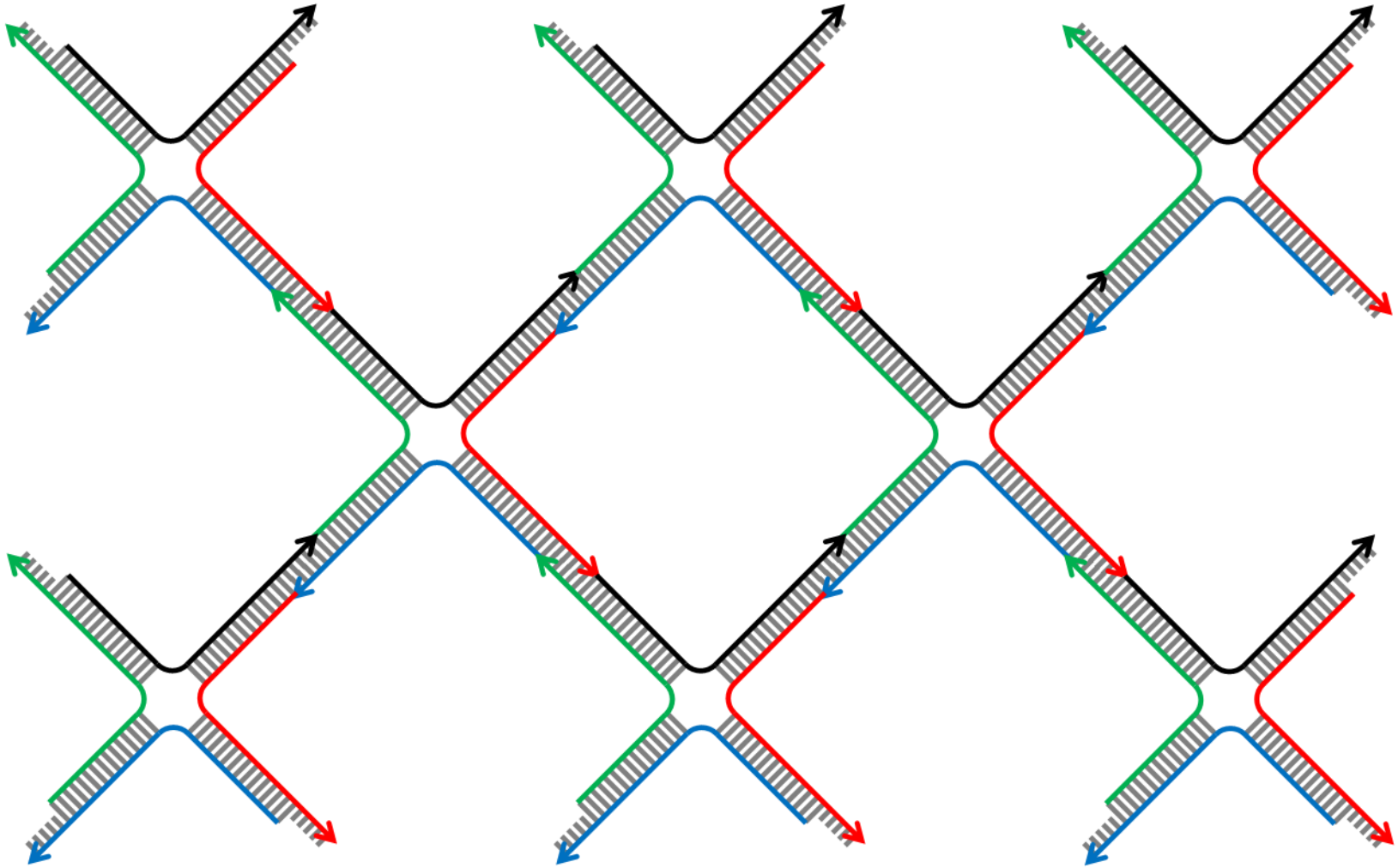

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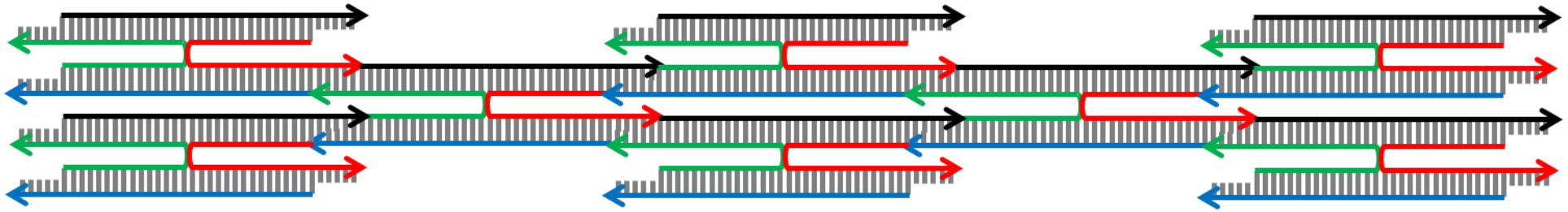

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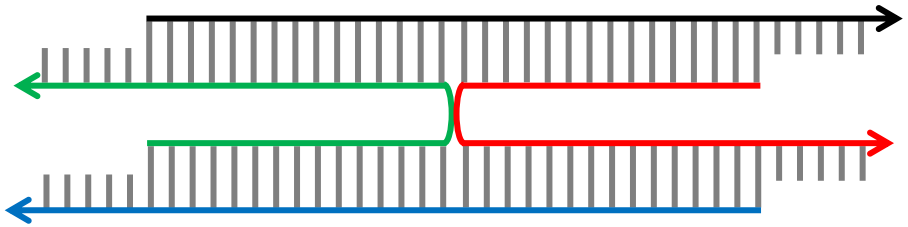
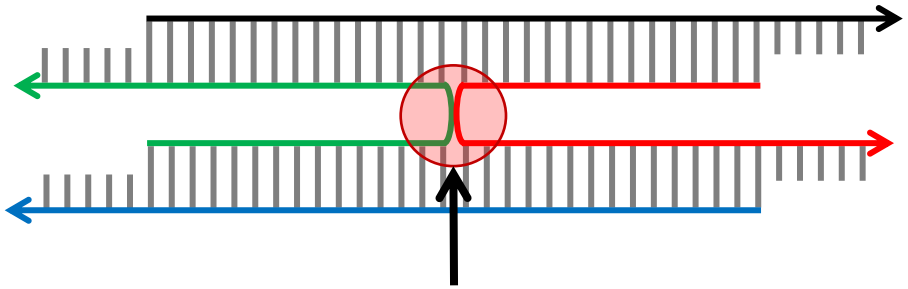
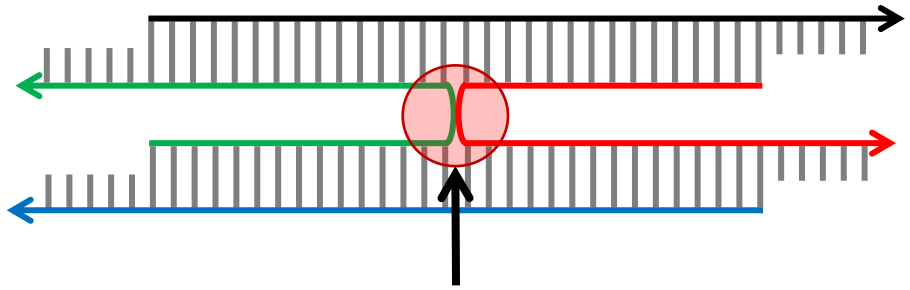


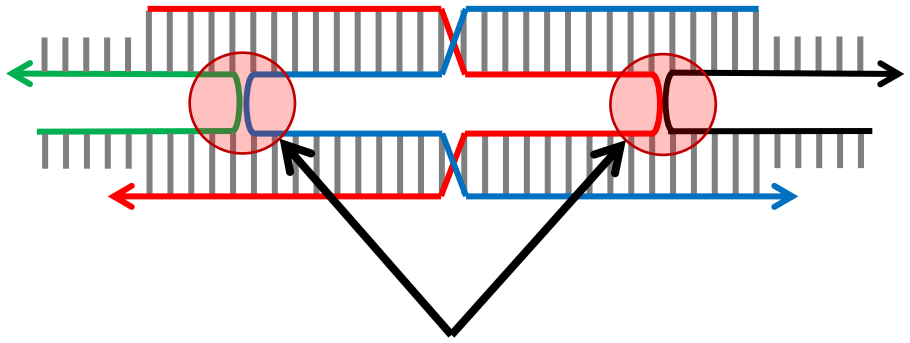
Practice of DNA tile self-assembly



Practice of DNA tile self-assembly



single *crossover*



double crossover

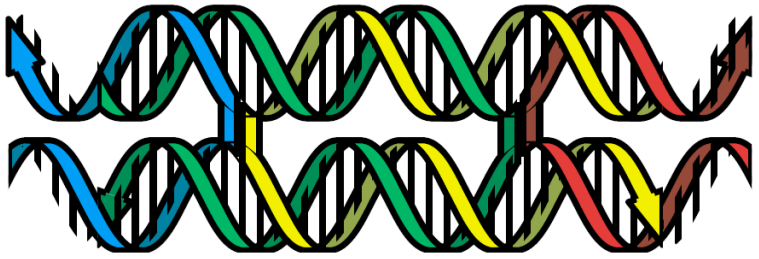
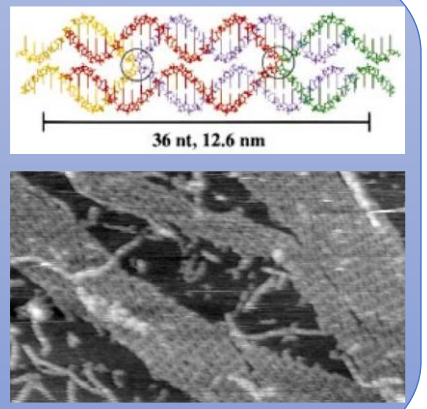


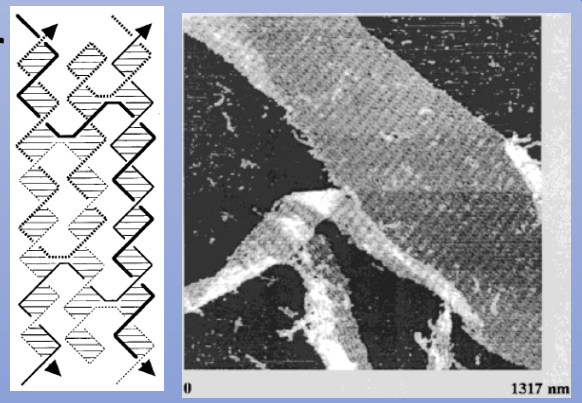
Figure from Schulman, Winfree, *PNAS* 2009

Practice of DNA tile self-assembly

double-crossover tile (Winfree, Liu, Wenzler, Seeman, *Nature* 1998)

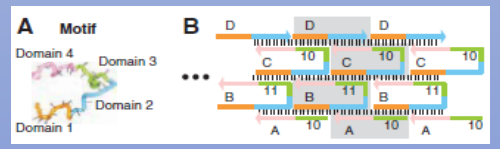


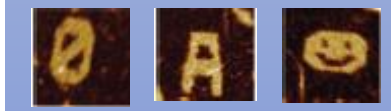
triple-crossover tile (LaBean, Yan, Kopatsch, Liu, Winfree, Reif, Seeman, *JACS* 2000)



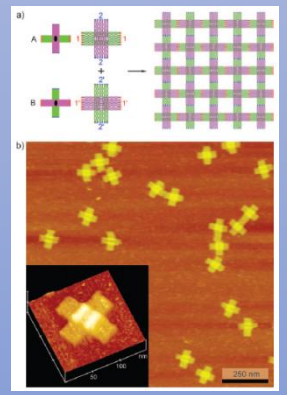
4x4 tile (Yan, Park, Finkelstein, Reif, LaBean, *Science* 2003)

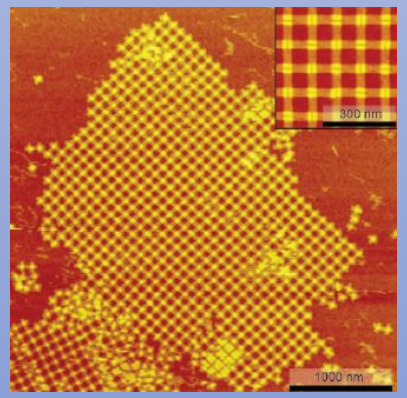
single-stranded tile (Yin, Hariadi, Sahu, Choi, Park, LaBean, Reif, *Science* 2008)



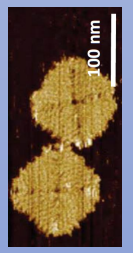


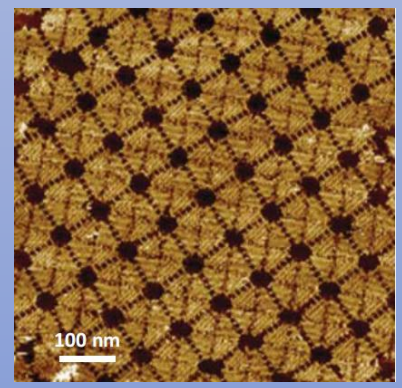
DNA origami tile (Liu, Zhong, Wang, Seeman, *Angewandte Chemie* 2011)





Tikhomirov, Petersen, Qian, *Nature Nanotechnology* 2017



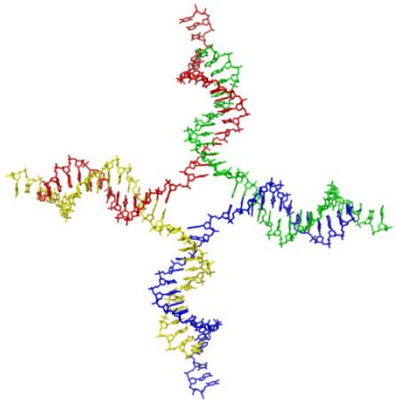


Theory of *algorithmic* self-assembly

What if…
… there is more than one tile type?
… some sticky ends are “weak”?

Erik Winfree

abstract Tile Assembly Model (aTAM)



Erik Winfree, Ph.D. thesis,
Caltech 1998

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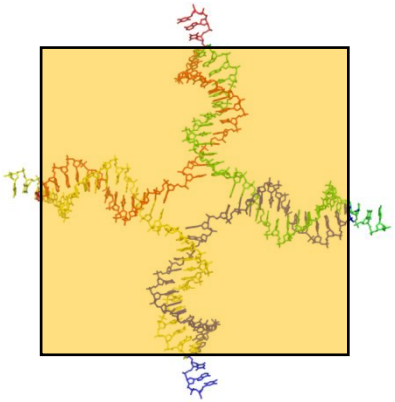
- **tile type** = unit square



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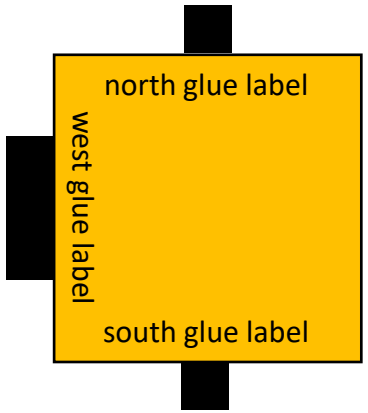
- **tile type** = unit square
- each side has a **glue** with a **label** and **strength** (0, 1, or 2)



strength 0

strength 1 (weak)

strength 2 (strong)

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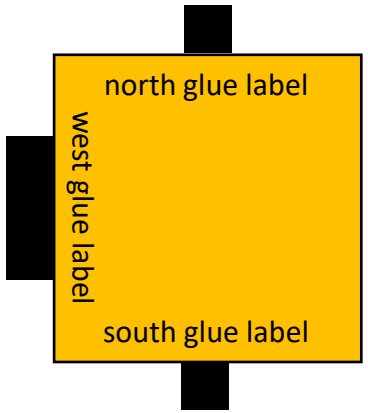

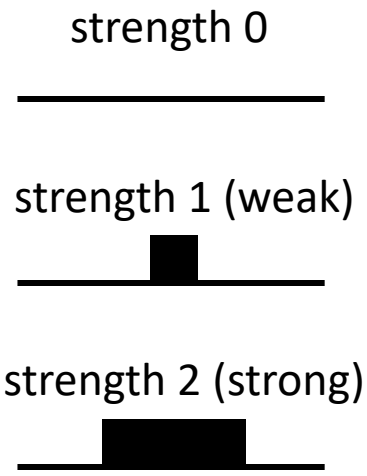

- finitely many tile **types**
- infinitely many **tiles**: copies of each type

Erik Winfree, Ph.D. thesis, Caltech 1998

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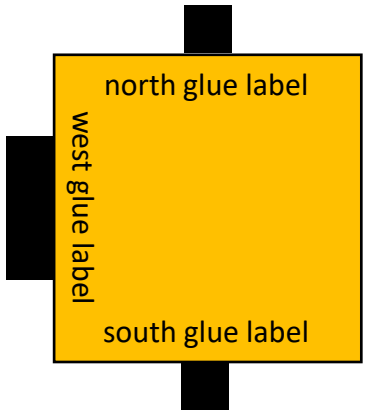

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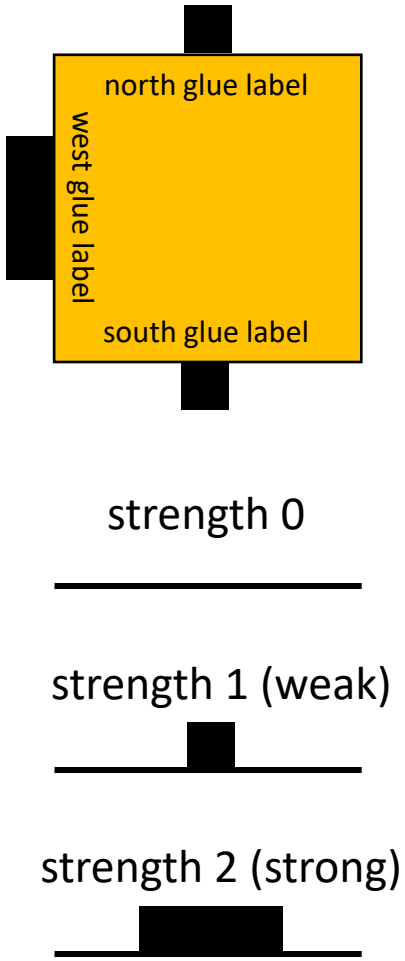
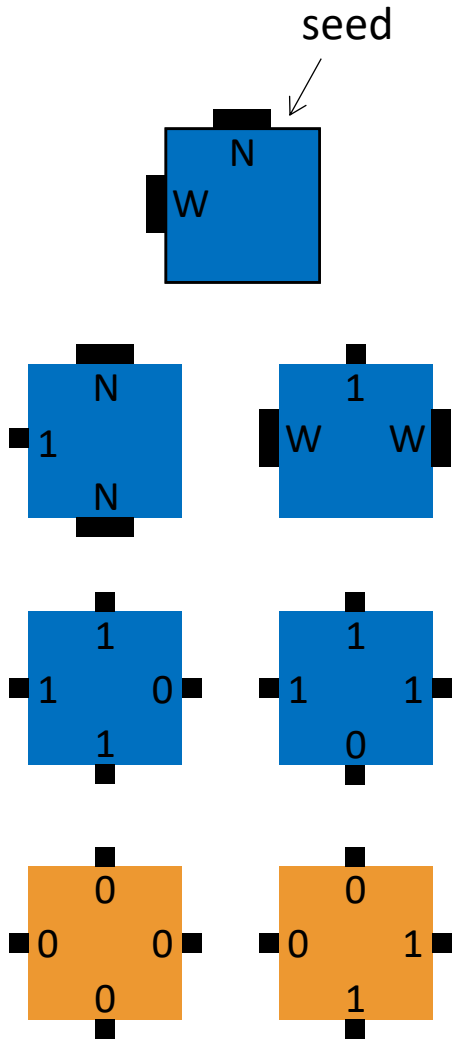


- finitely many tile **types**
- infinitely many **tiles**: copies of each type
- assembly starts as a single copy of a special **seed** tile
- tile can bind to the assembly if total binding strength ≥ 2 (two weak glues or one strong glue)

Erik Winfree, Ph.D. thesis, Caltech 1998

Example tile set



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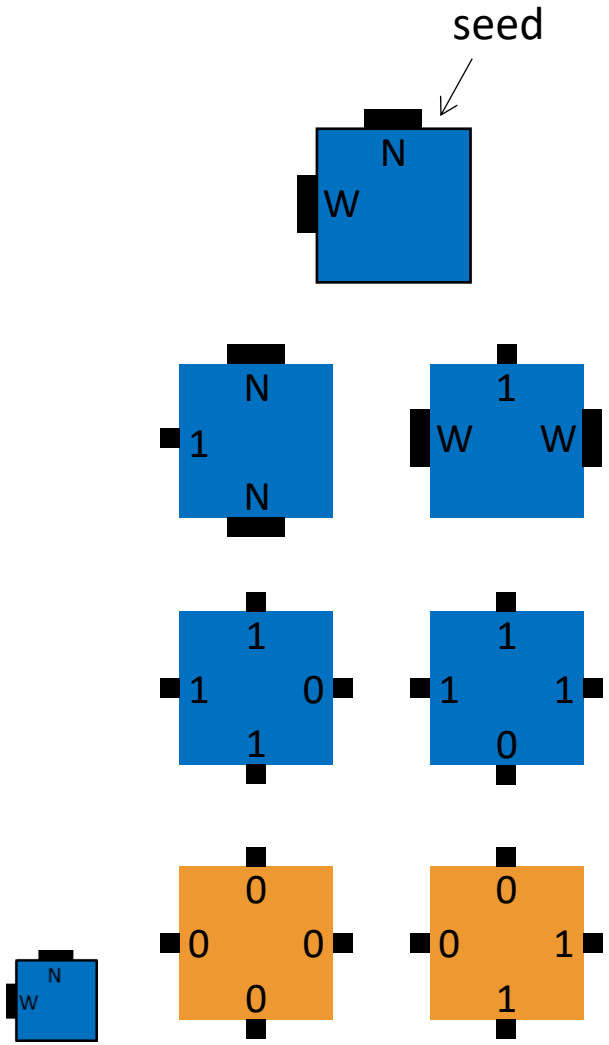
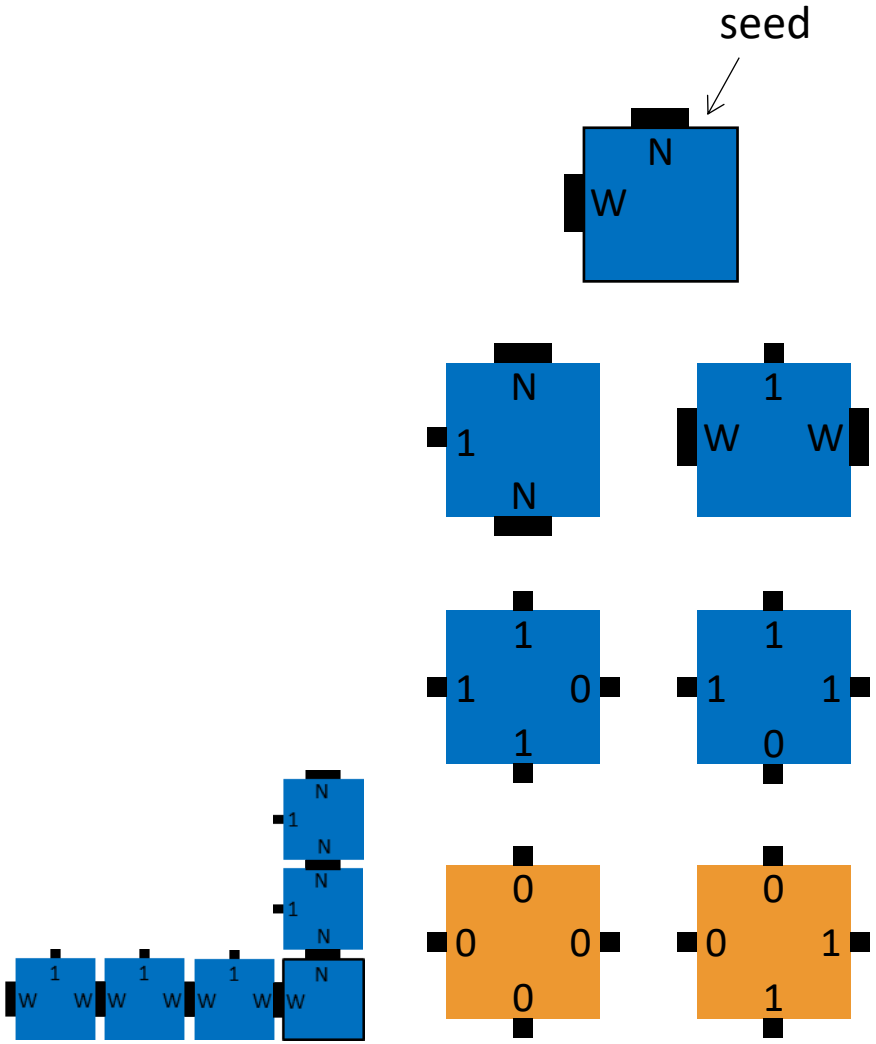
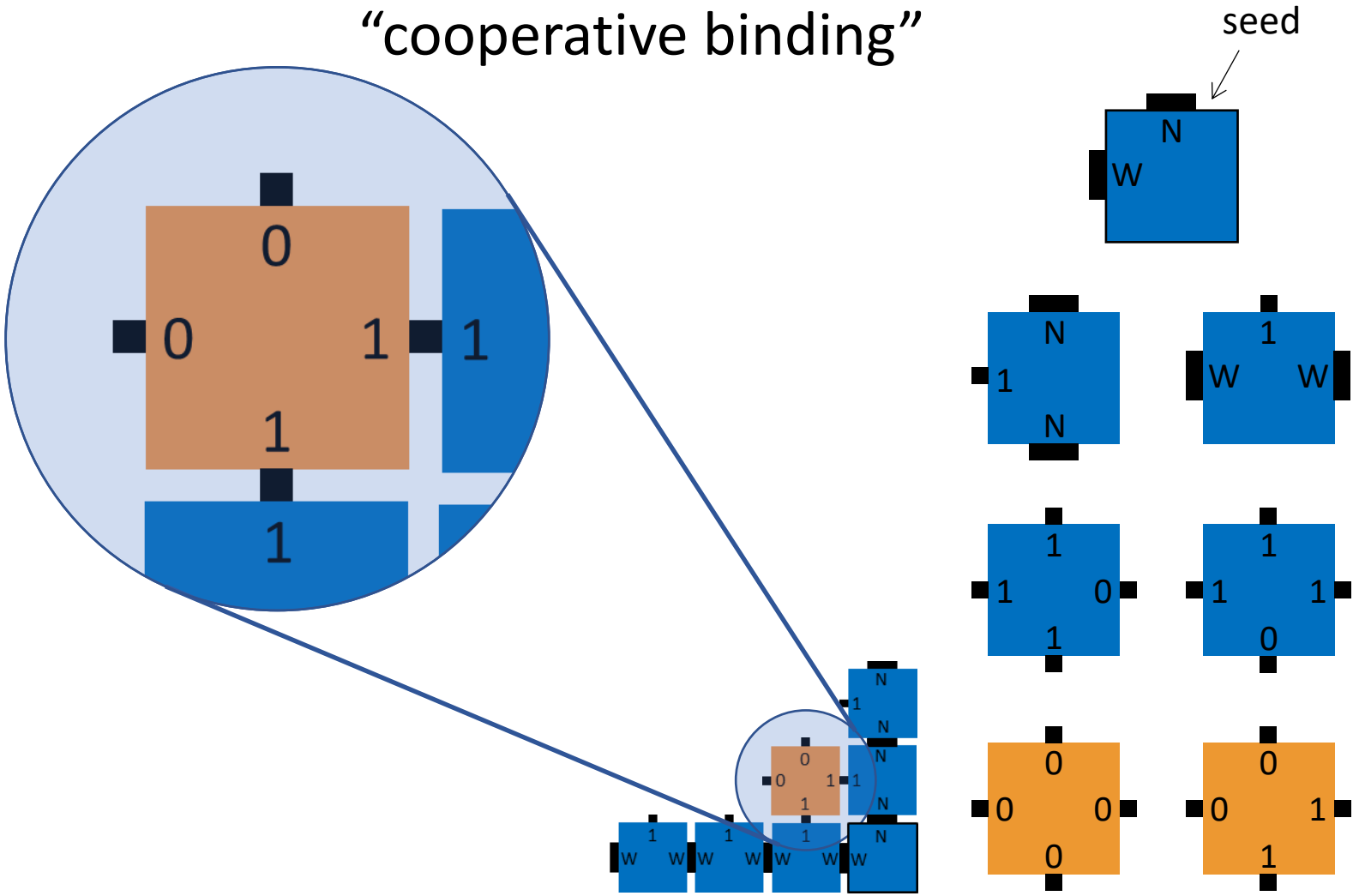


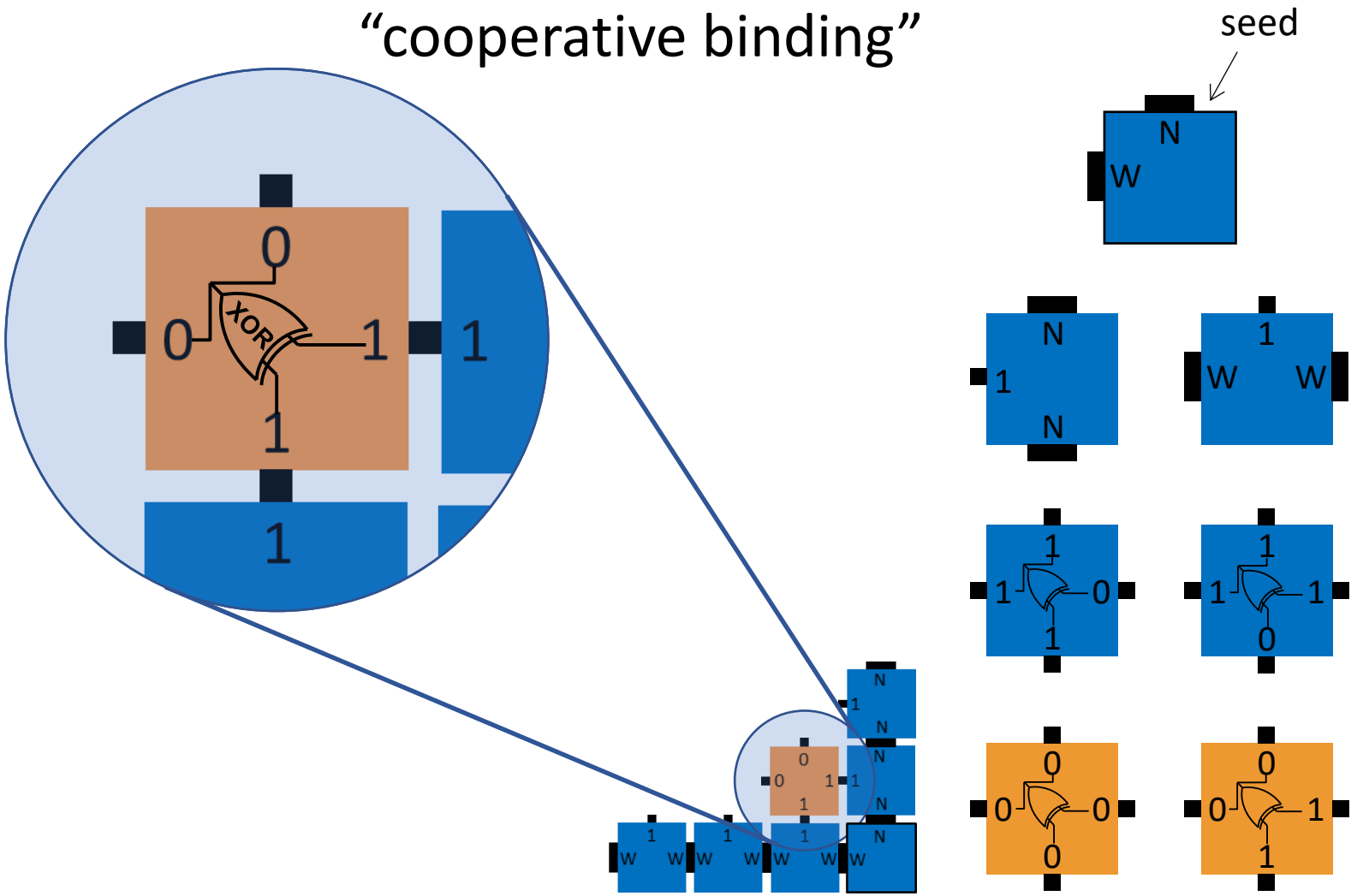
Example tile set



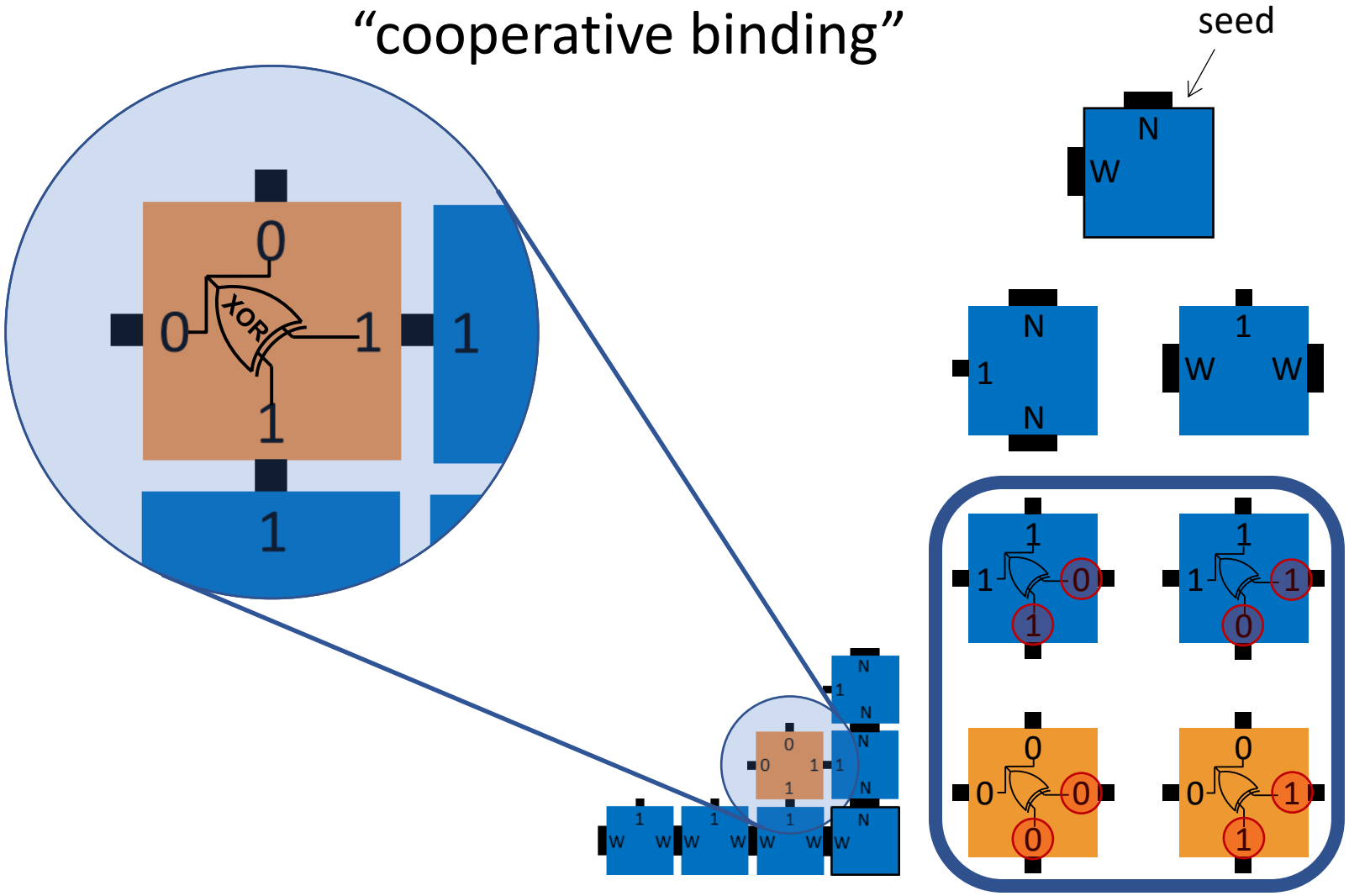
Example tile set



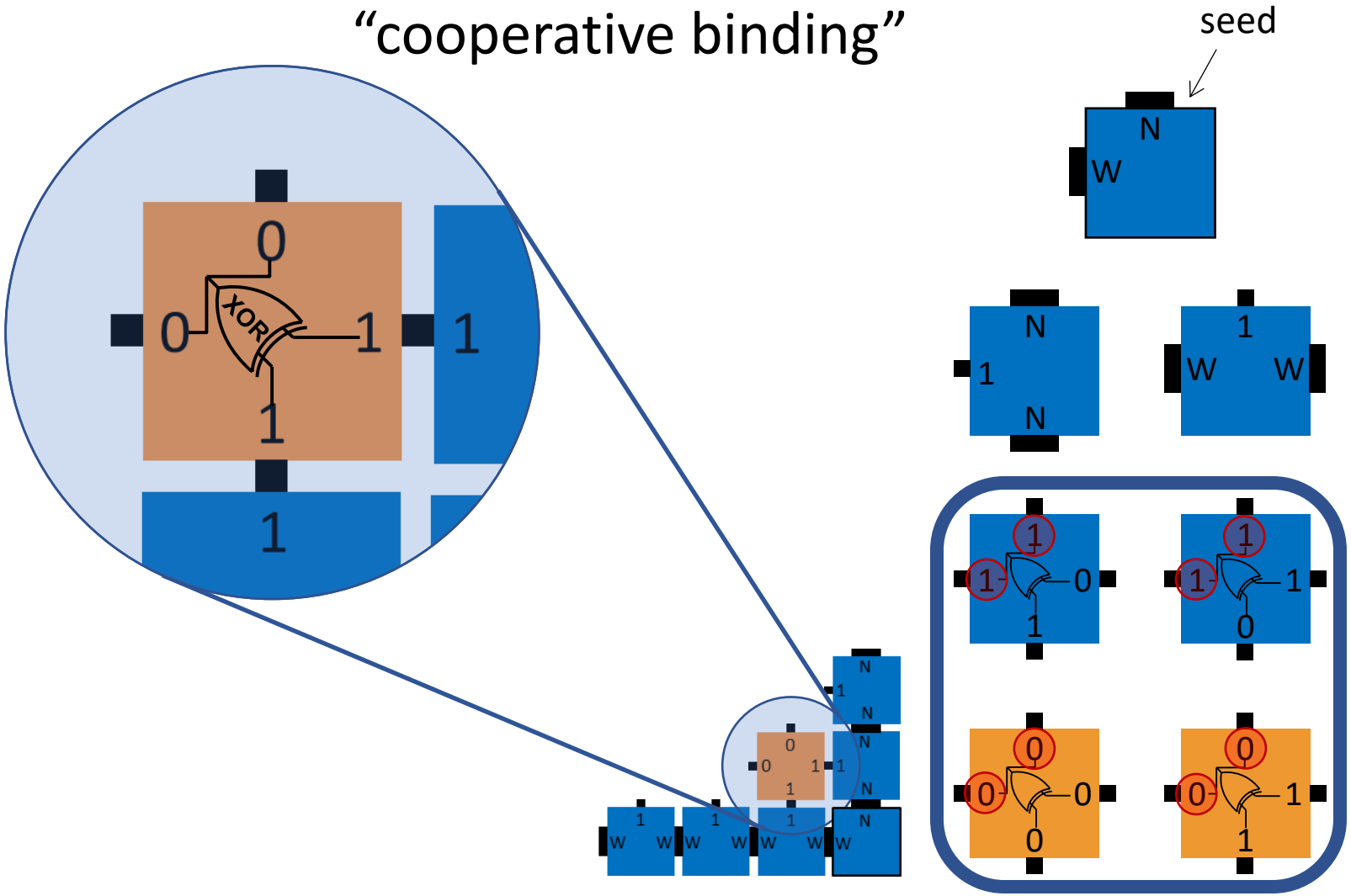
Example tile set



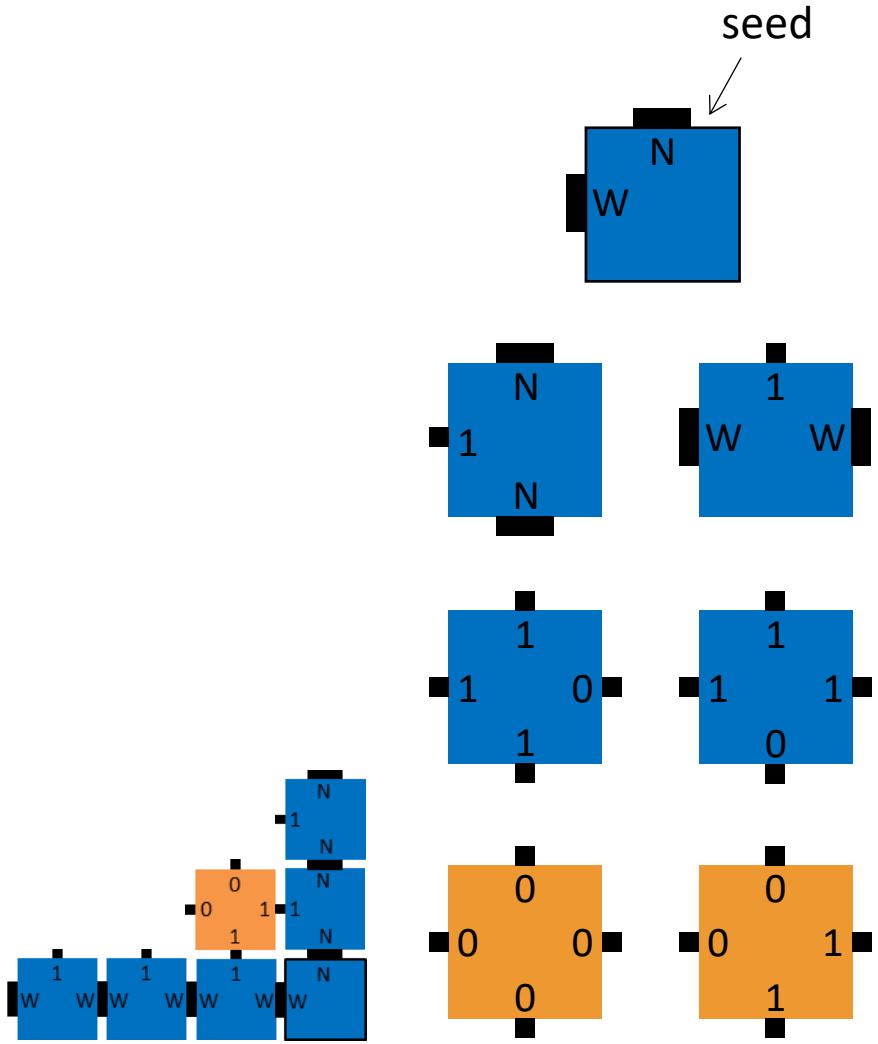
Example tile set

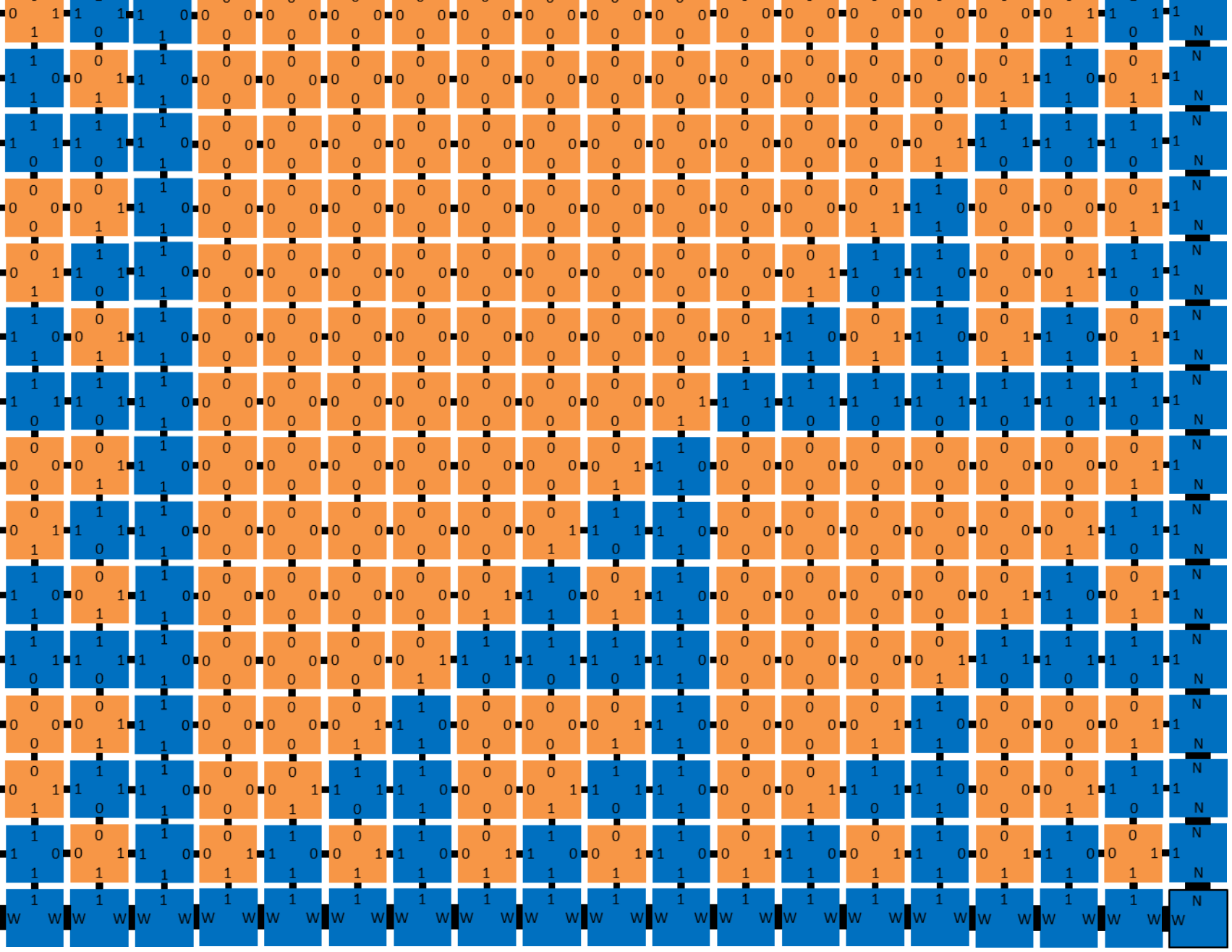


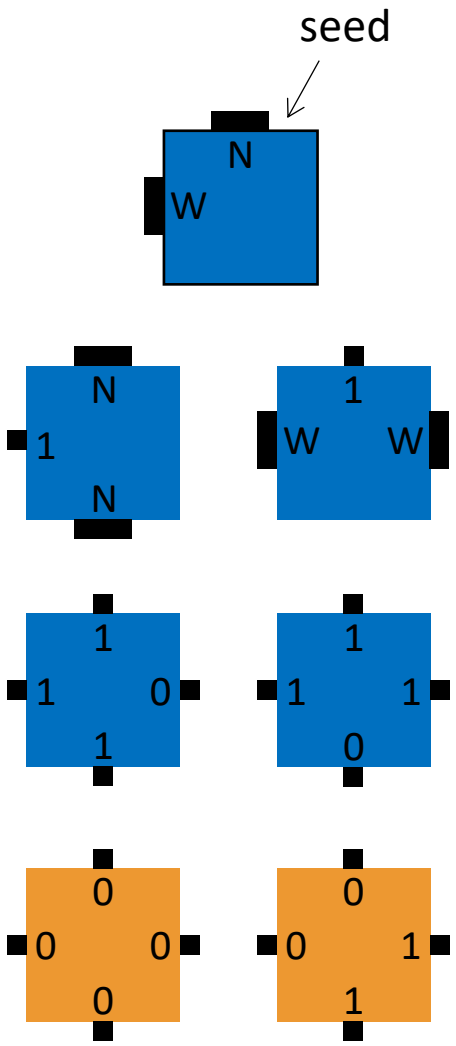
Example tile set

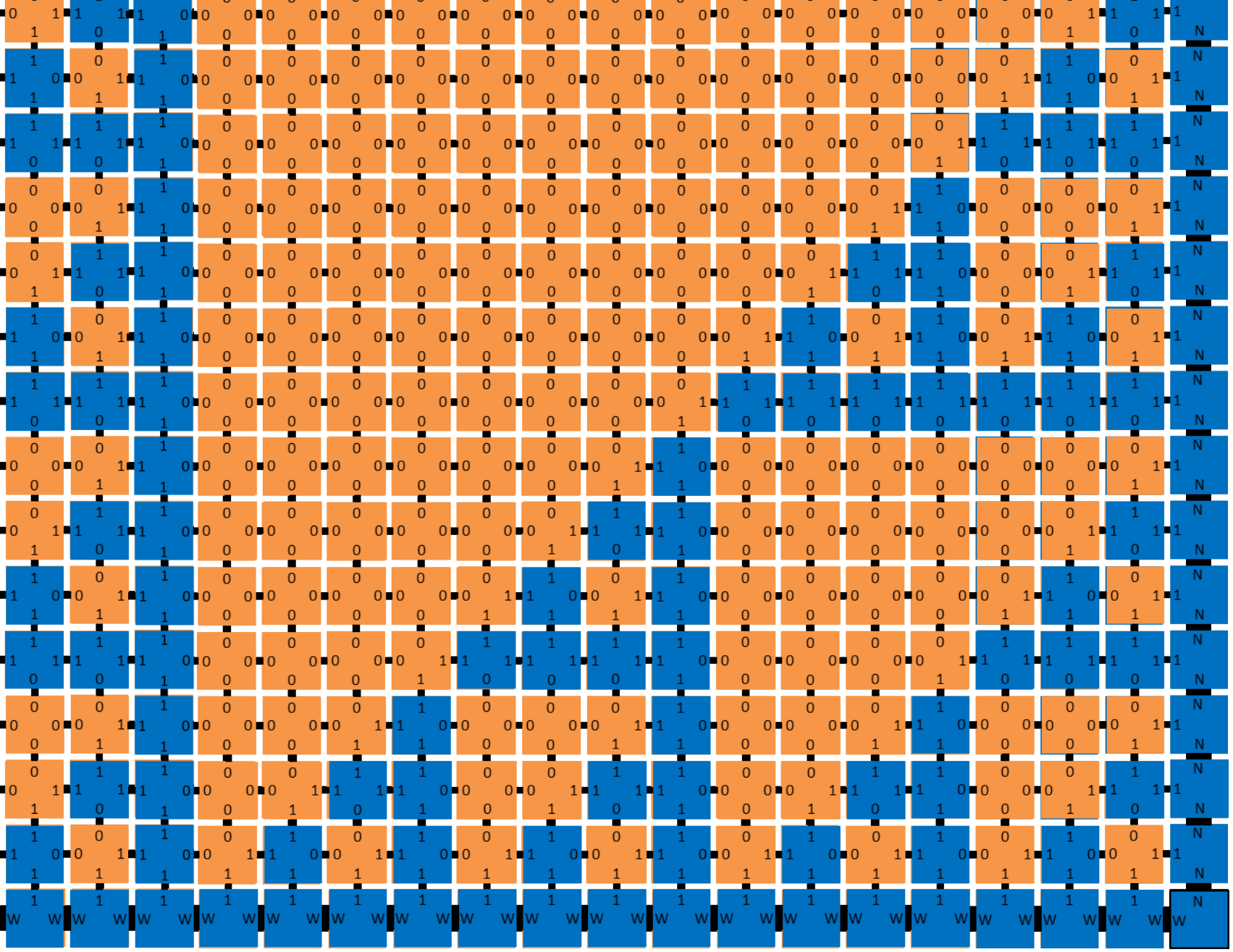


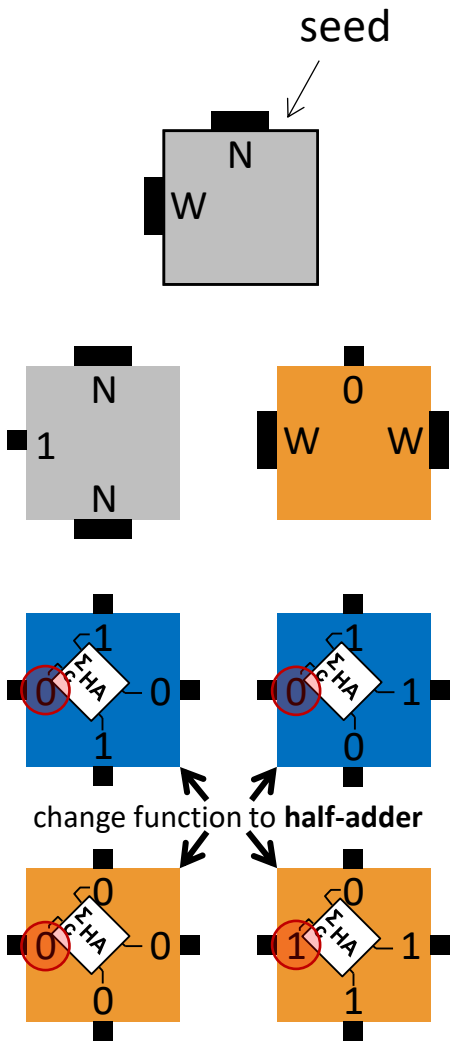
Example tile set

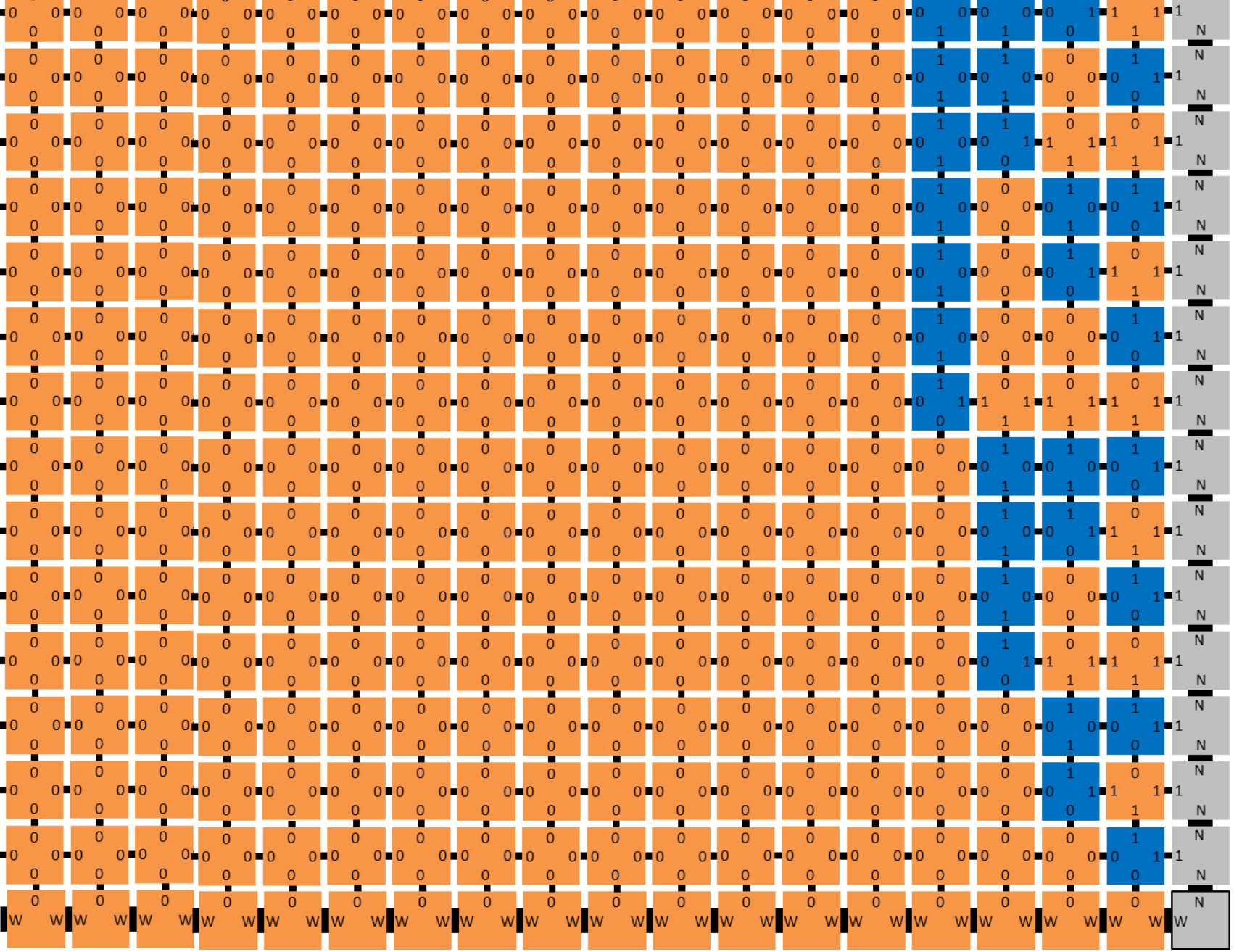


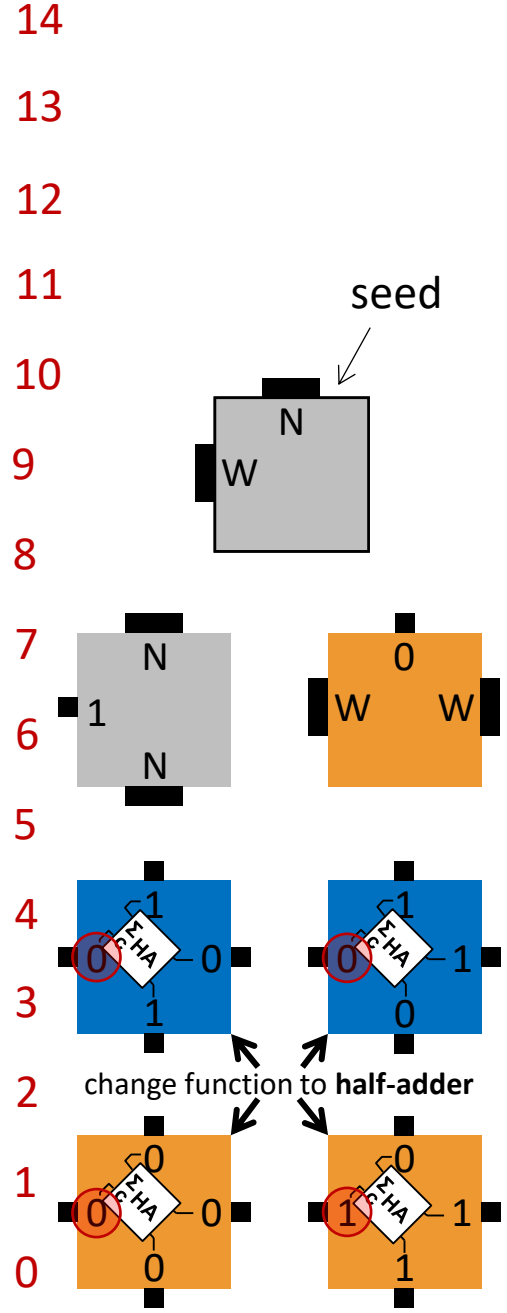



seed
N
W
N
1
N
1
W
W
1
1
0
1
1
1
0
0
0
0
0
0
1

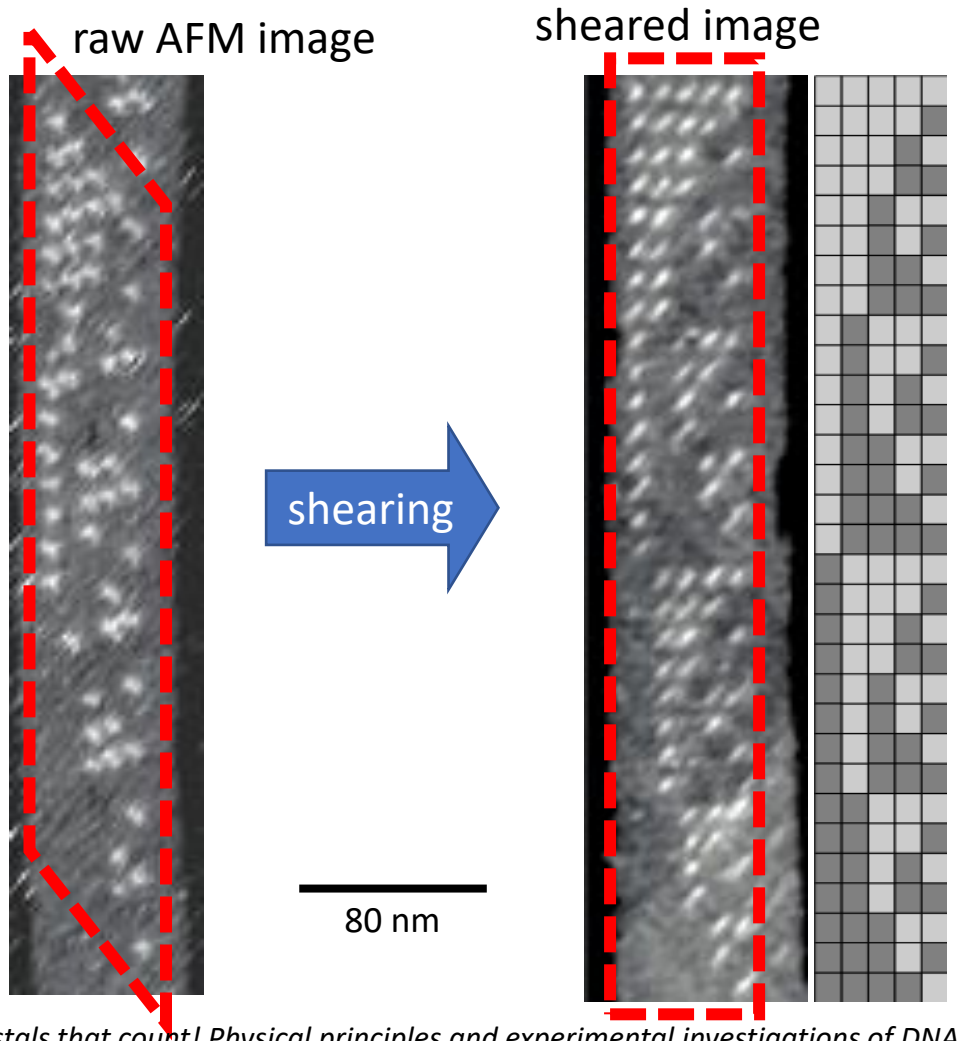



seed
N
W
N
1
N
0
W
W
1
0
ΣHA
1
0
1
0
ΣHA
1
1
0
change function to **half-adder**
0
0
ΣHA
0
0
0
1
ΣHA
1
1
1




seed
N
W
N
1
N
0
W
W
1
0
1
0
1
0
1
0
change function to half-adder
0
0
0
0
0
1
0
1

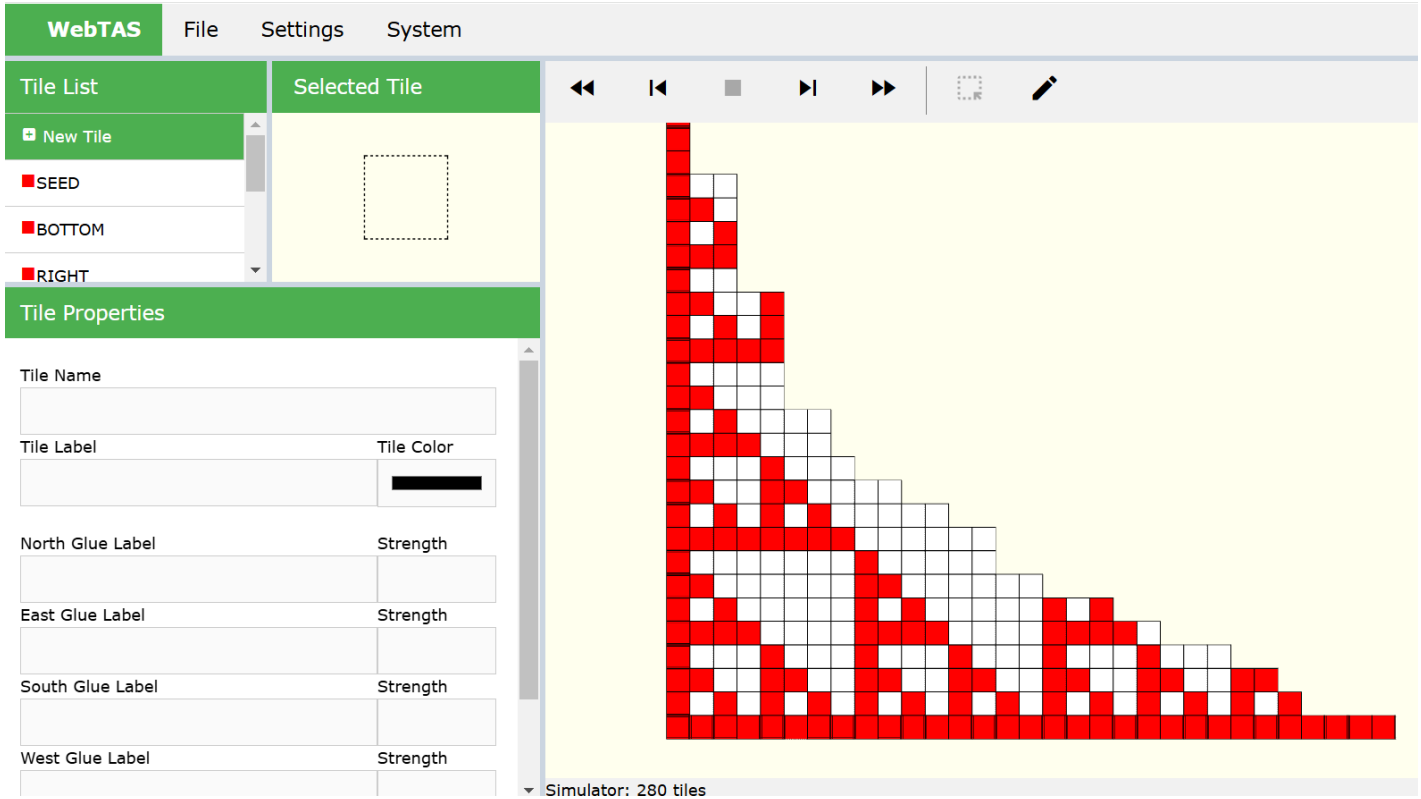
Algorithmic self-assembly in action



[*Crystals that count! Physical principles and experimental investigations of DNA tile self-assembly*, Constantine Evans, Ph.D. thesis, Caltech, 2014]

aTAM simulator (WebTAS by Daniel Hader)

http://self-assembly.net/software/WebTAS/WebTAS-latest/



Tip: for editing tile types, I find it much easier to edit the text files directly than to use the GUI, which is tedious. You may also consider writing code to generate the files.

Xgrow by Constantine Evans: https://github.com/DNA-and-Natural-Algorithms-Group/xgrow
older xgrow (by Erik Winfree) https://www.dna.caltech.edu/Xgrow/

Tile complexity of squares

Tile complexity

- Resource bound to minimize, like time or memory with a traditional algorithm.

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- We start with *n* x *n* squares as the “simplest” benchmark shape.
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- [*Note: we have not formally defined the aTAM yet… first let’s build intuition.*]

The program size complexity of self-assembled squares

Question: How many tile types do we need to self-assemble an *n* x *n* square?

https://www.dna.caltech.edu/Papers/squares_STOC.pdf
This paper is directly responsible for convincing many theoretical computer scientists that DNA self-assembly is worth studying.

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Concretely: how to assemble a 4 x 4 square?

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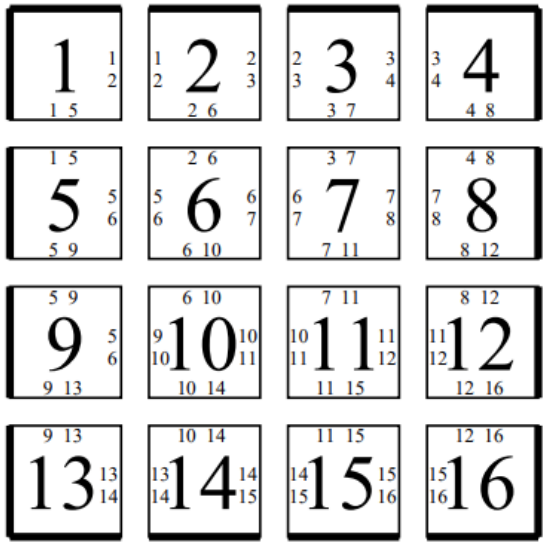

All glues are strength 2
(alternately: all are strength 1 and *temperature* $\tau = 1$)

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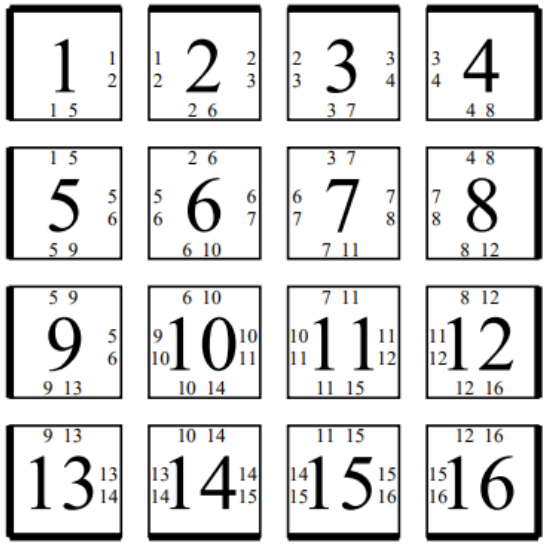

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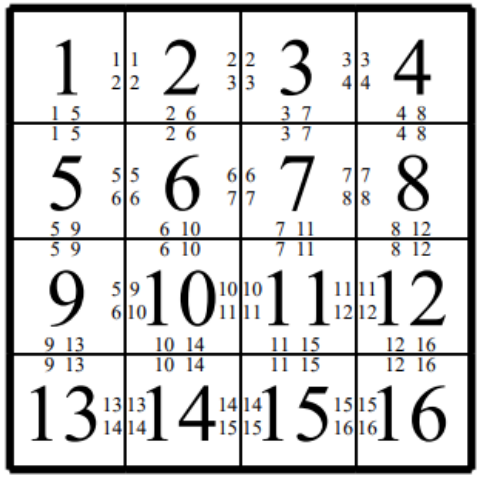

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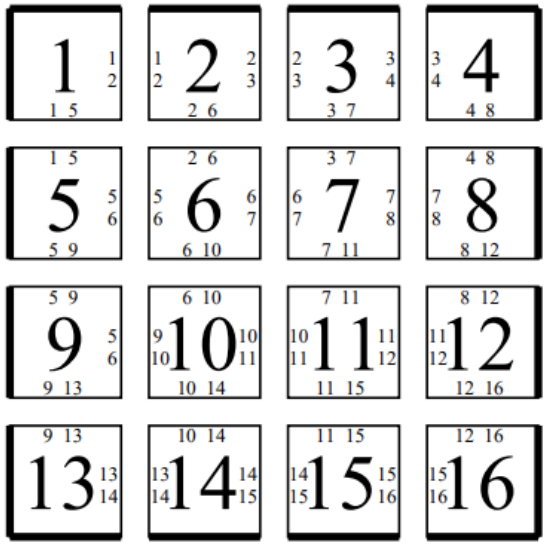

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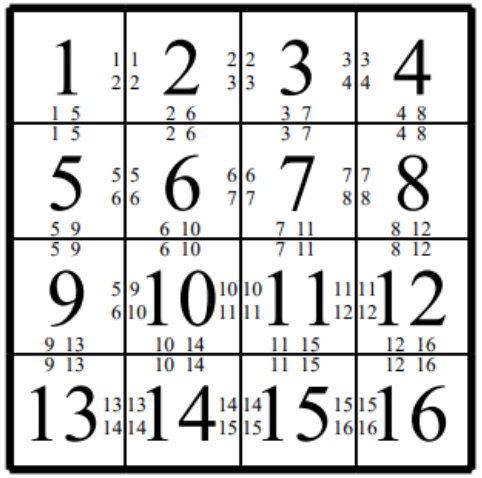

How many tile types does this construction need in general to assemble an *n* x *n* square?

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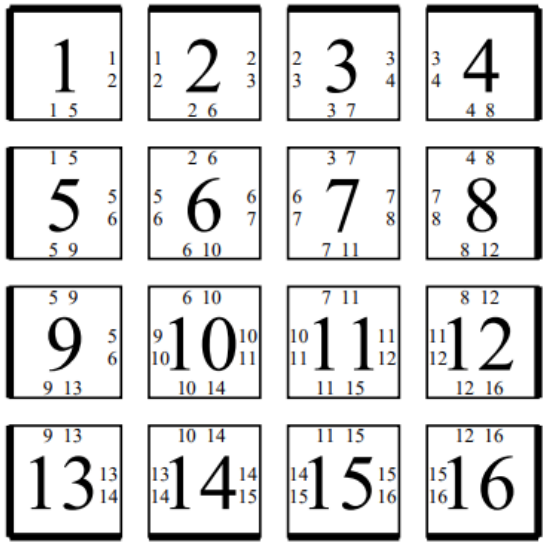
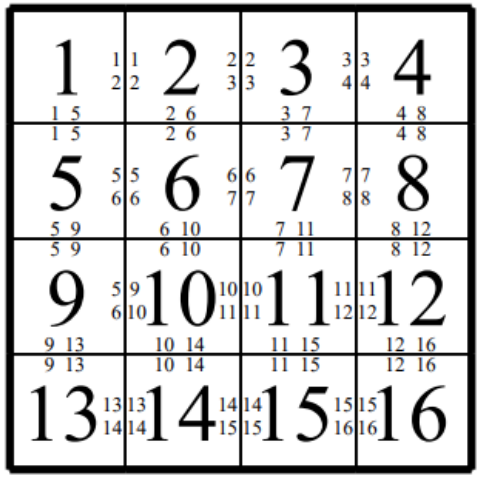


All glues are strength 2
(alternately: all are strength 1 and *temperature* $\tau = 1$)



How many tile types does this construction need in general to assemble an *n* x *n* square?

n^2

https://www.dna.caltech.edu/Papers/squares_STOC.pdf
This paper is directly responsible for convincing many theoretical computer scientists that DNA self-assembly is worth studying.

Tile complexity at temperature $\tau = 1$ (i.e., no cooperative binding allowed)

Is n^2 optimal?
Can we do better?

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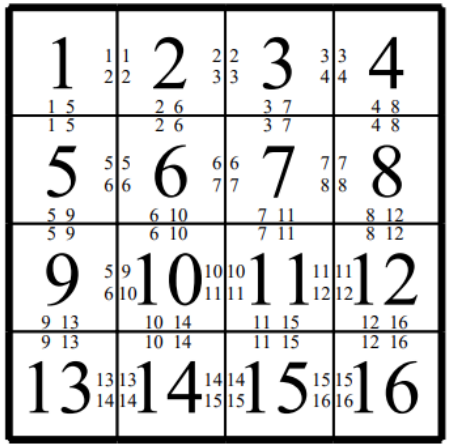

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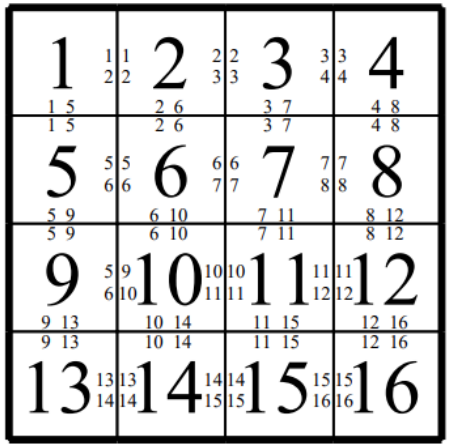

Theorem: At temperature $\tau = 1$, if all pairs of adjacent tiles bind with positive strength, then for every positive integer n, n^2 tile types are <u>necessary</u> to self-assemble an n x n square.

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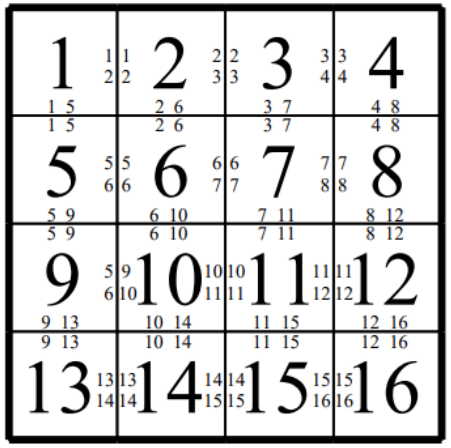
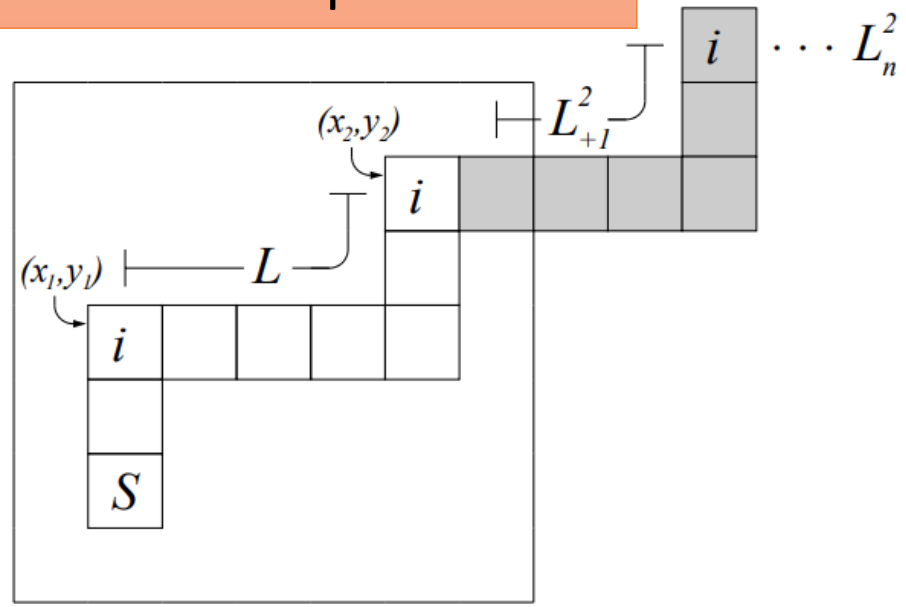


Theorem: At temperature $\tau = 1$, if all pairs of adjacent tiles bind with positive strength, then for every positive integer n, n^2 tile types are necessary to self-assemble an n x n square.

Proof: Suppose for contradiction we use the same tile type i at positions (x_1,y_1) and (x_2,y_2). Then they have a path L between them with all positive-strength glues, and this can happen instead:



Tile complexity at temperature $\tau = 1$, where not all adjacent tiles are bound

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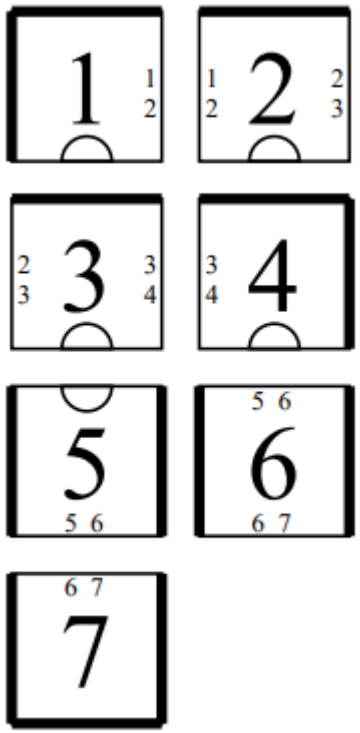

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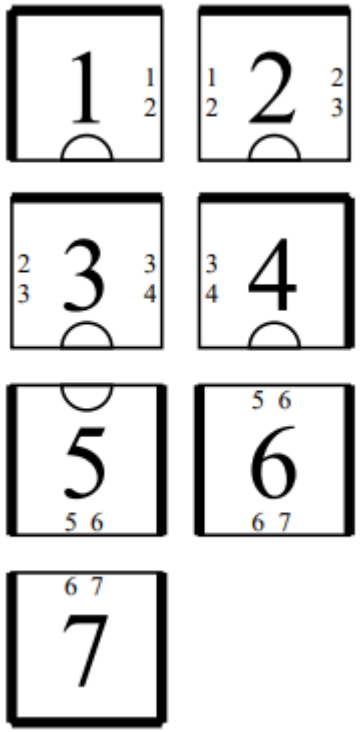

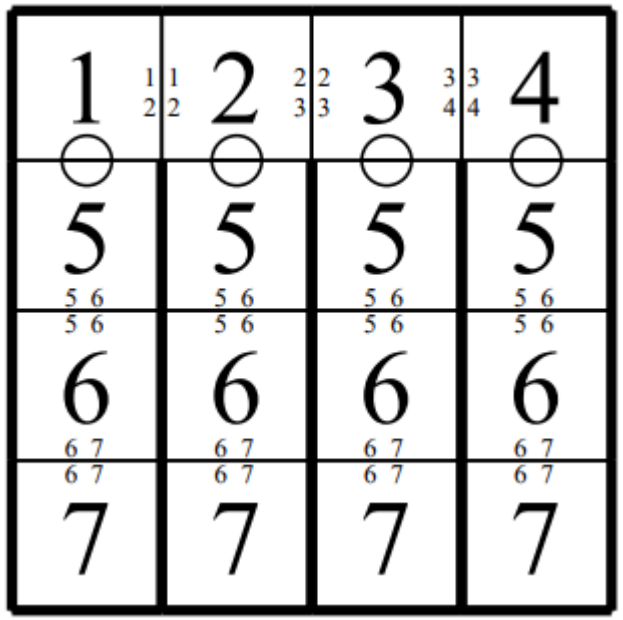

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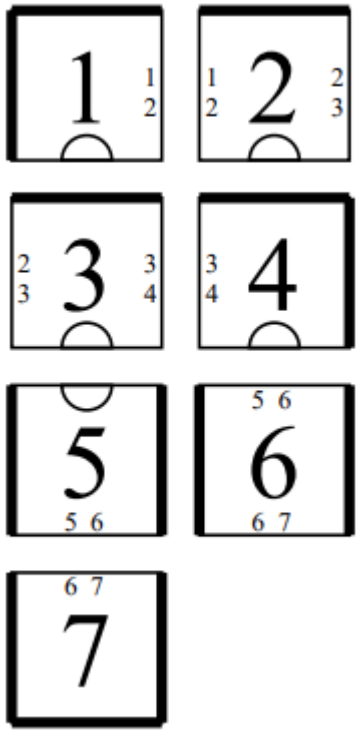

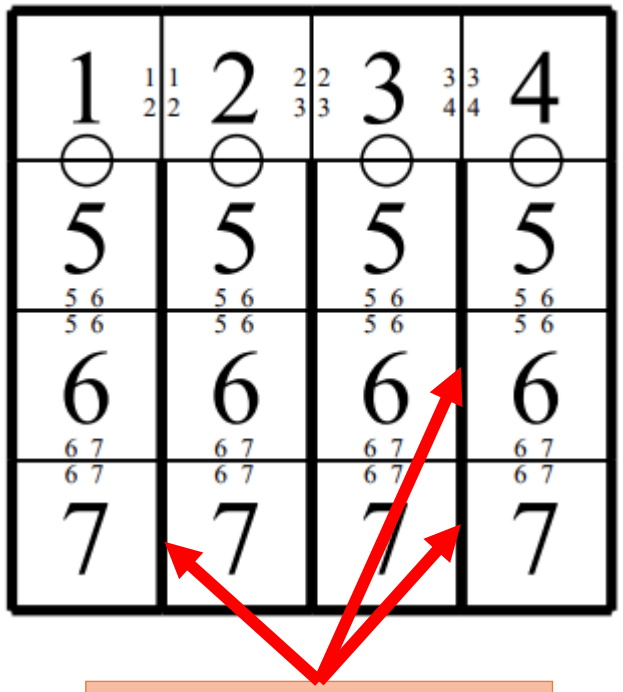

strength-0 glues

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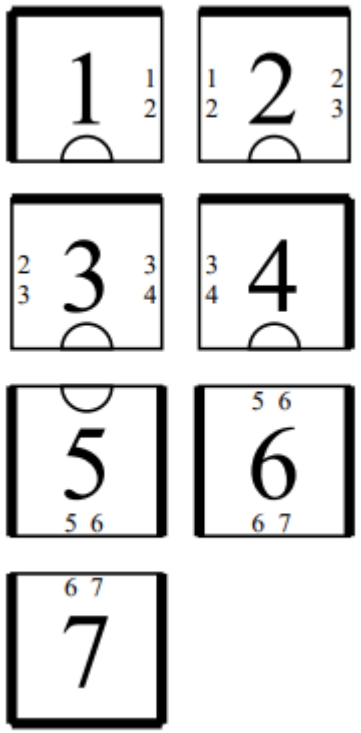

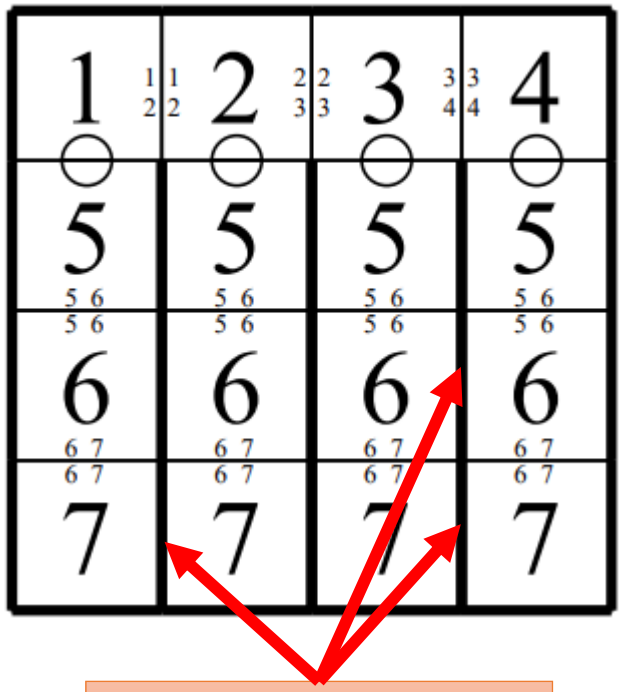

strength-0 glues

Tile complexity of this construction?

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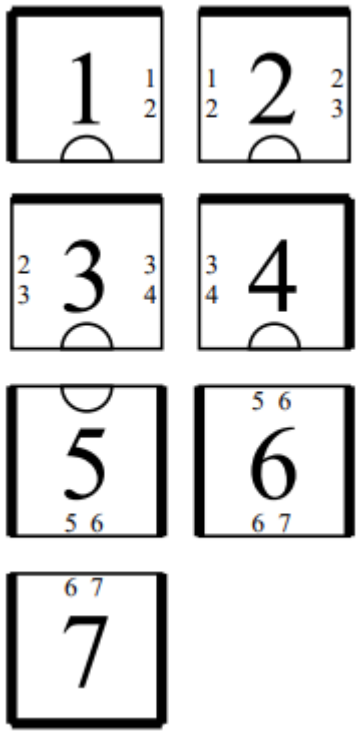

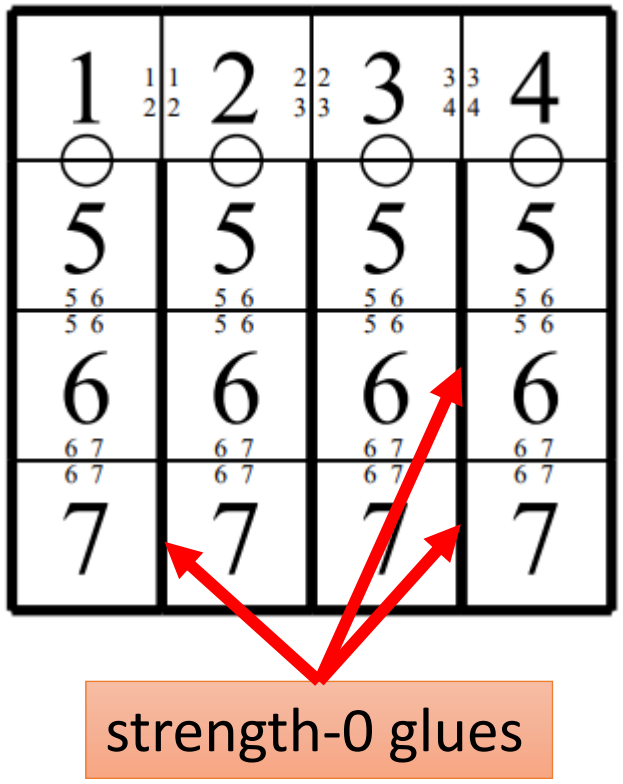

strength-0 glues

Tile complexity of this construction?

$2n - 1 = O(n)$

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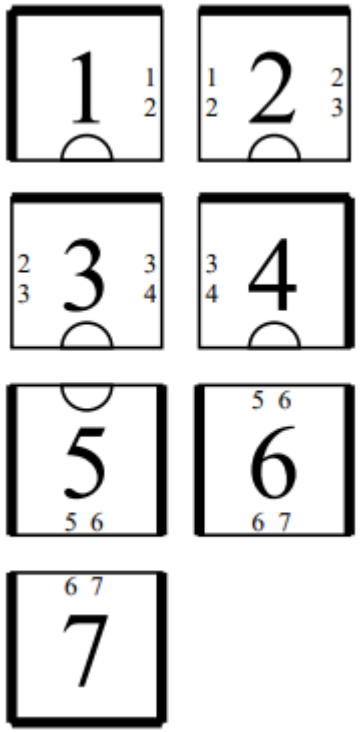

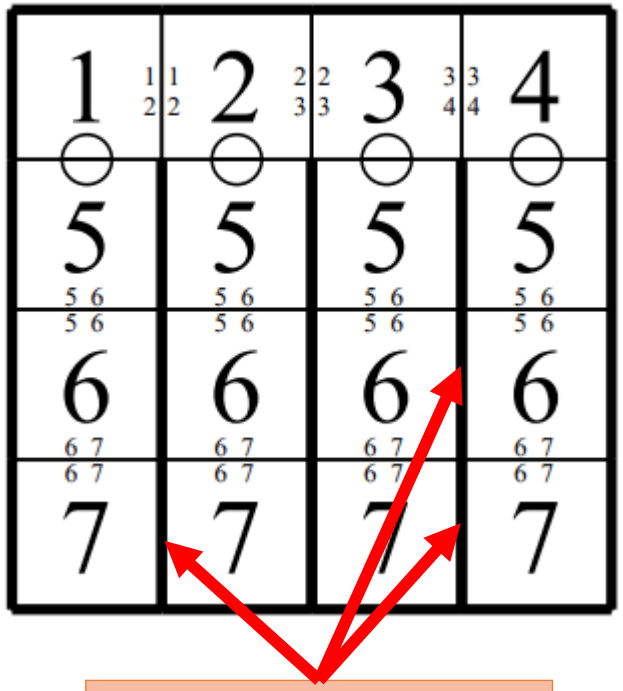

strength-0 glues

Tile complexity of this construction?

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Conjecture: The temperature $\tau = 1$ tile complexity of an n x n square is $\Omega(n)$.
(most recent progress:
https://arxiv.org/abs/1902.02253
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Tile complexity at temperature $\tau = 2$ (i.e., cooperative binding allowed)

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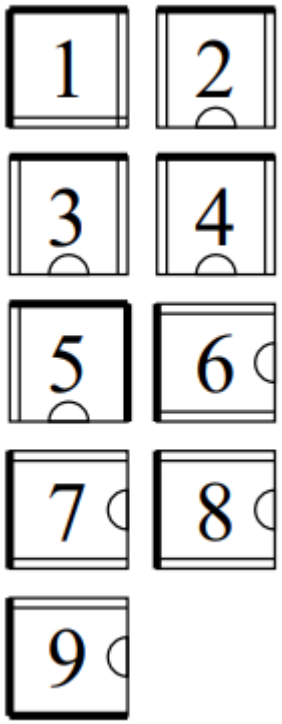

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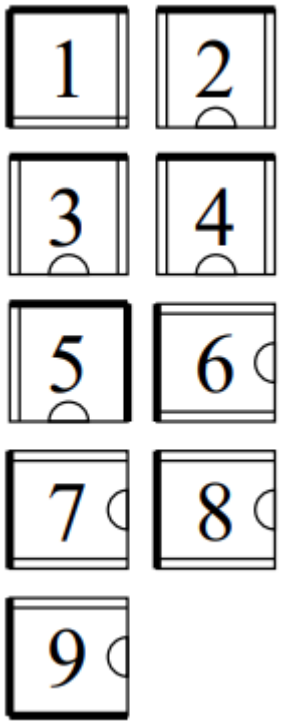

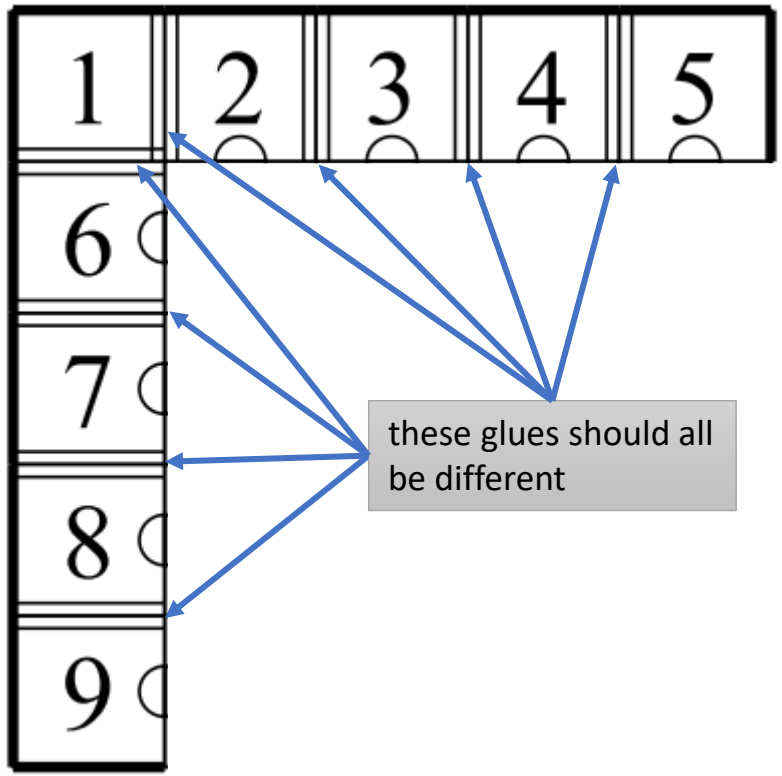

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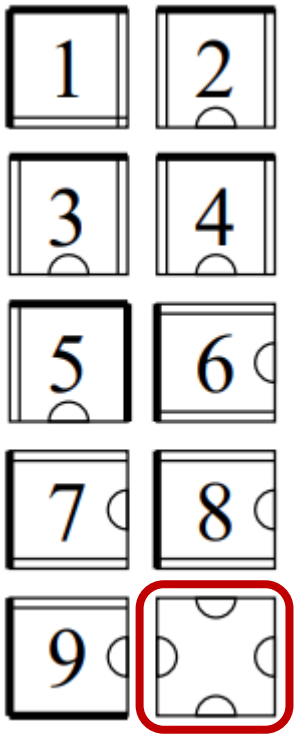

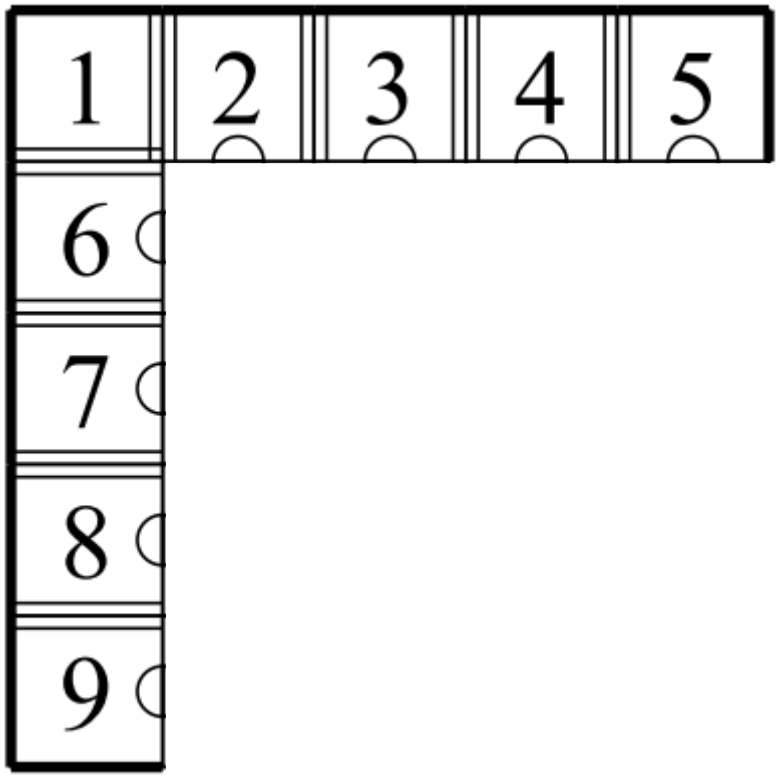

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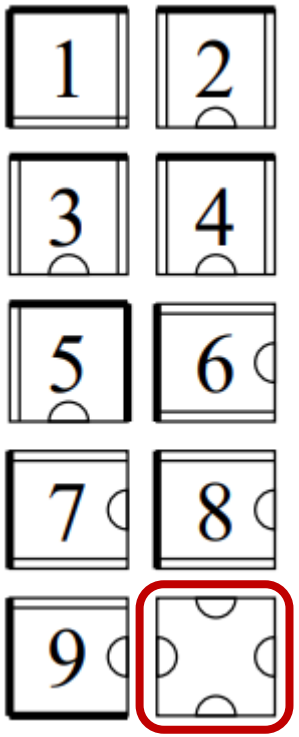

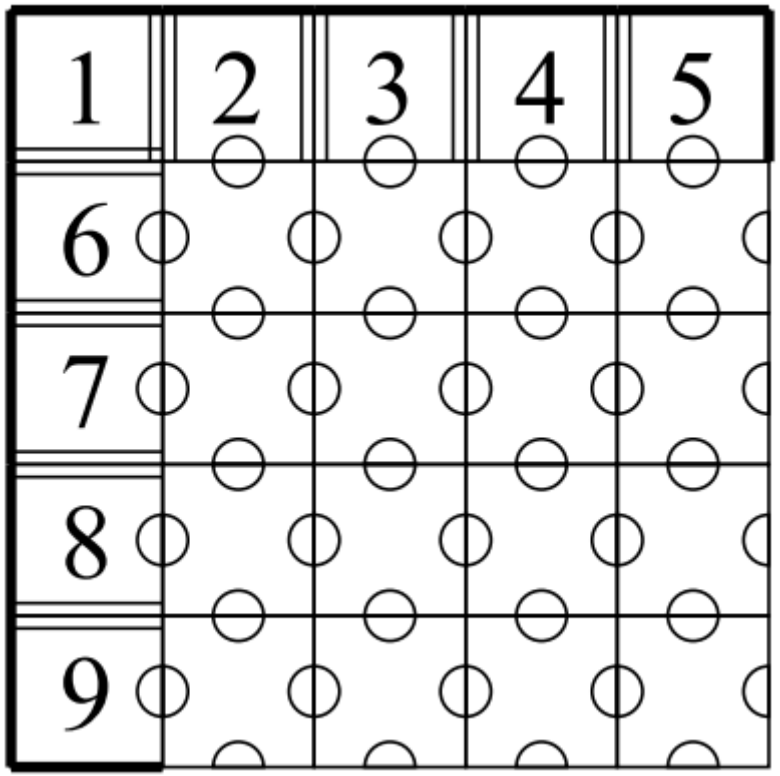

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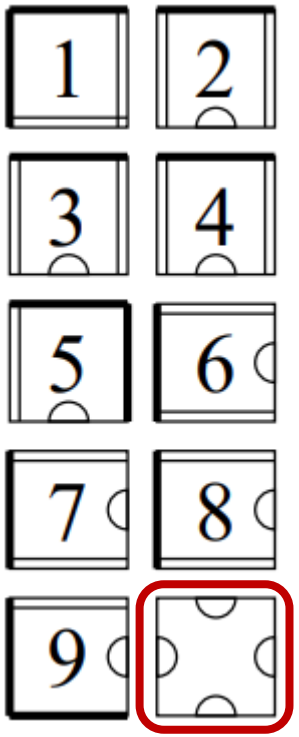

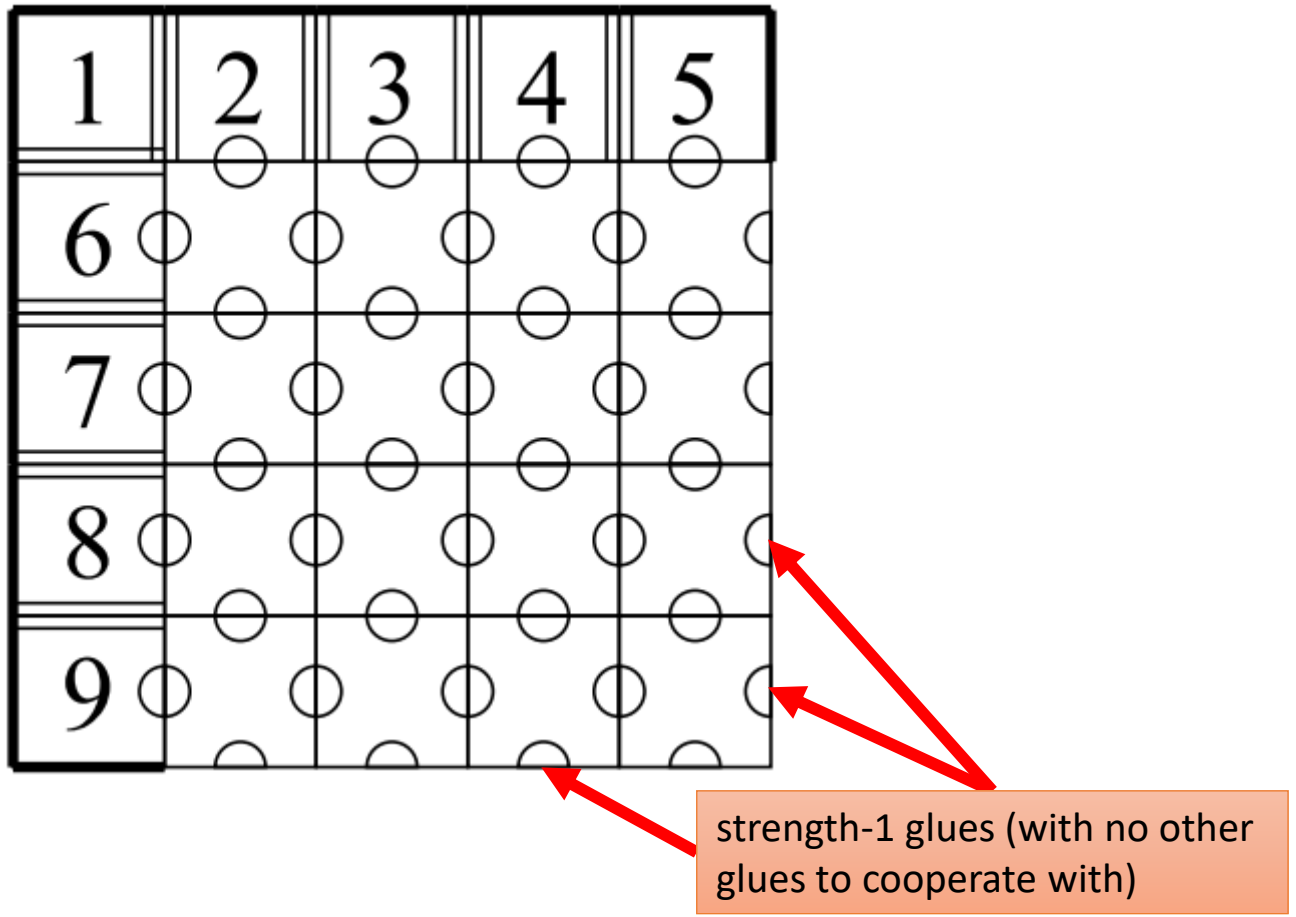

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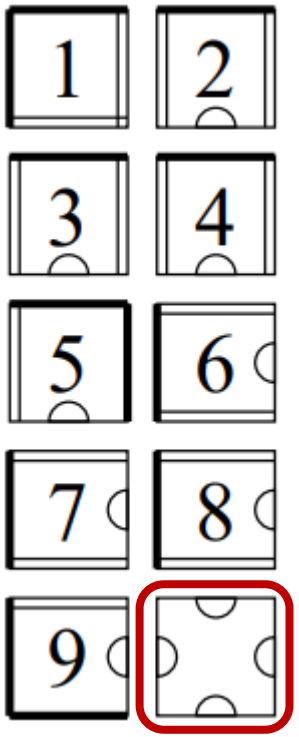

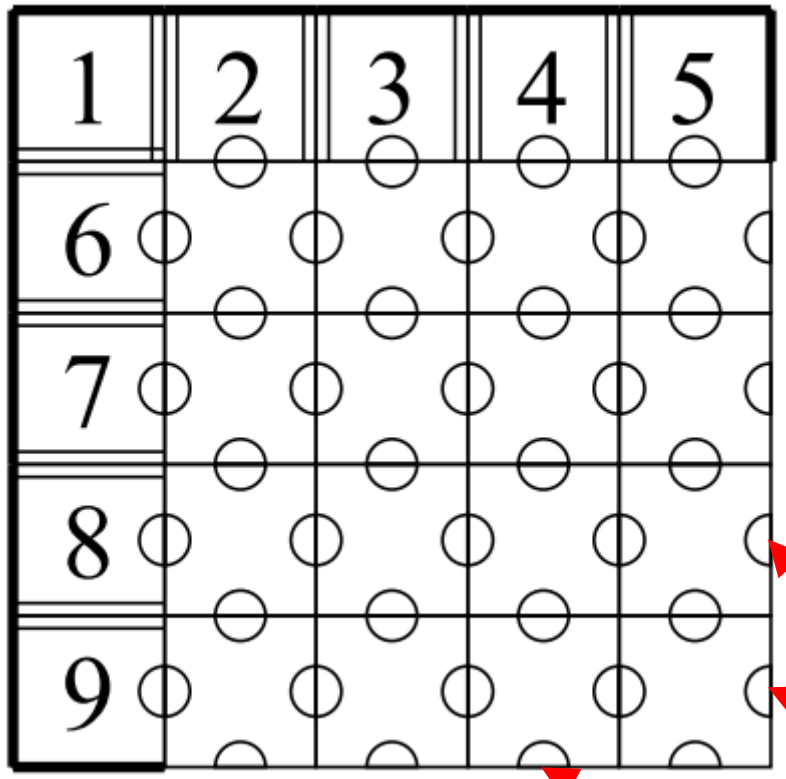

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strength-1 glues (with no other glues to cooperate with)

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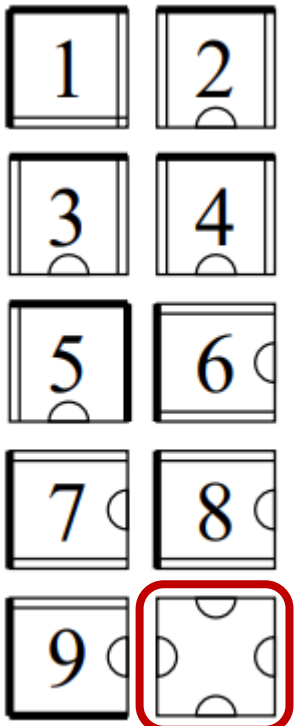
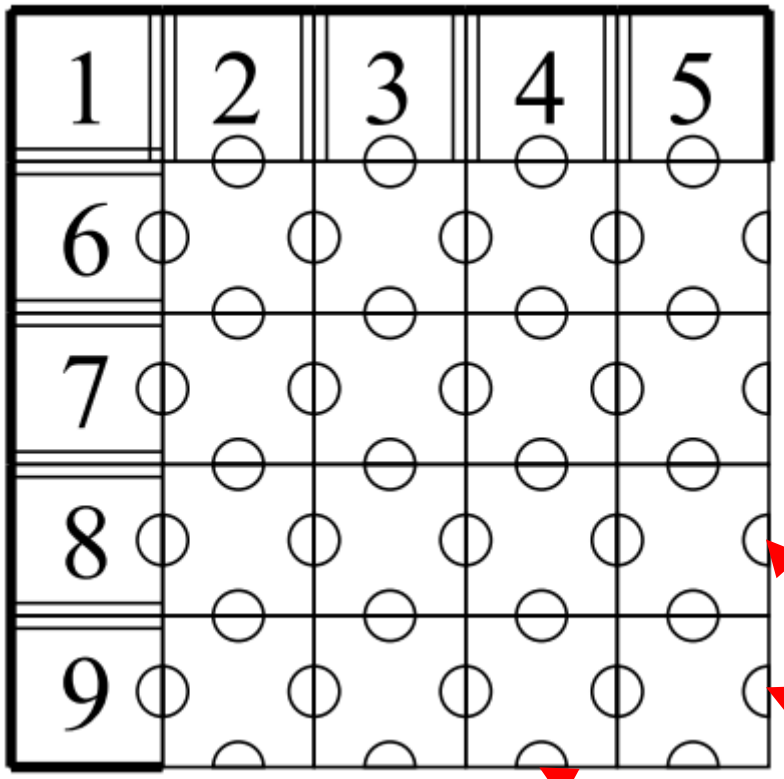


This tile completes an n x n "L shape" into an n x n square.



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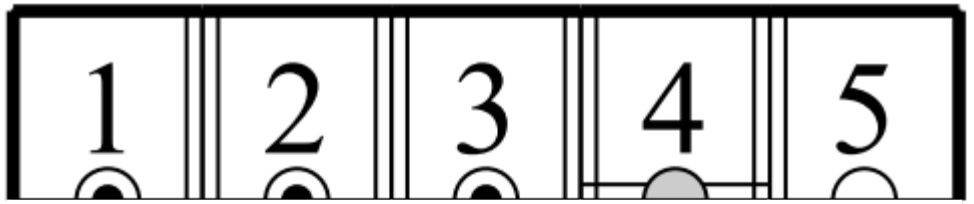
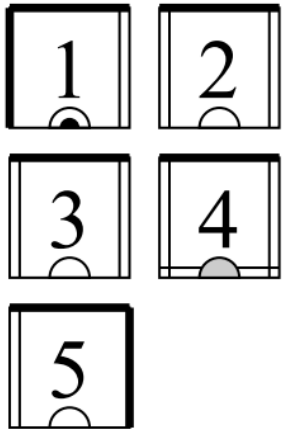
Tile complexity at temperature $\tau = 2$

Goal: complete a 1 x *n* line into an *n* x *n* square





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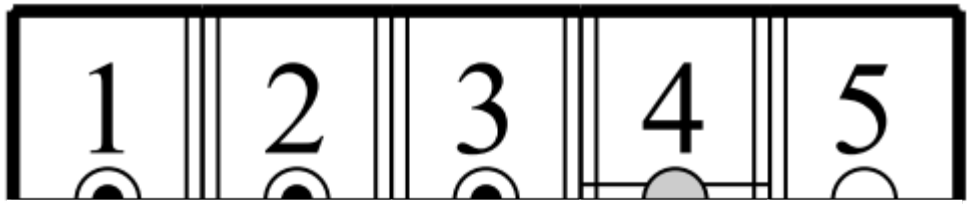

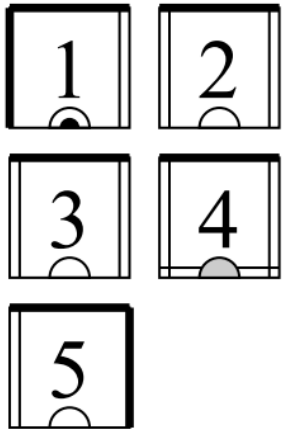

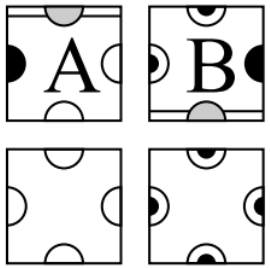

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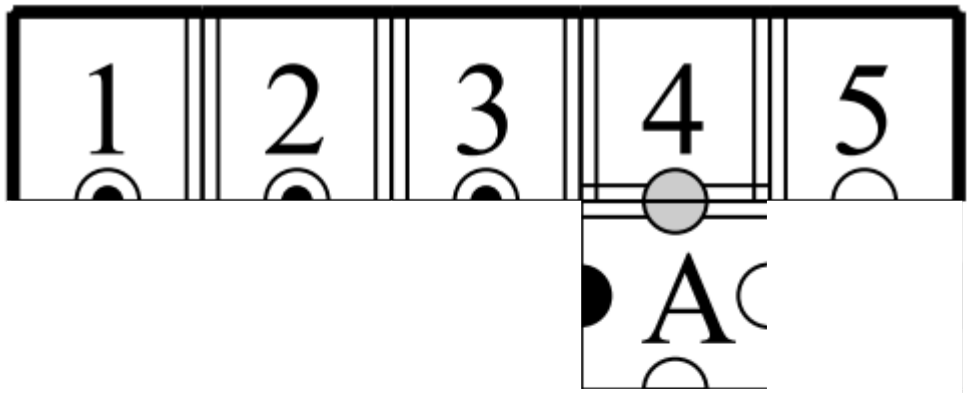

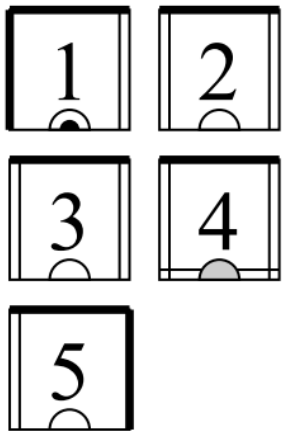

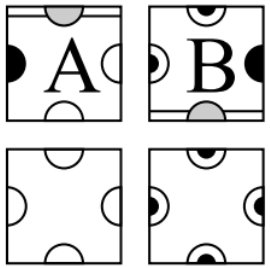

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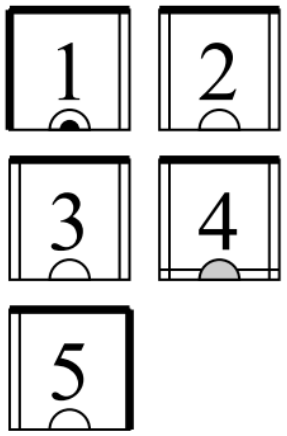

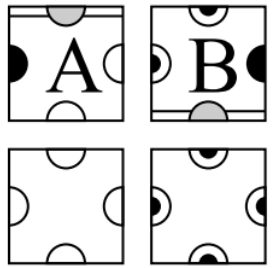

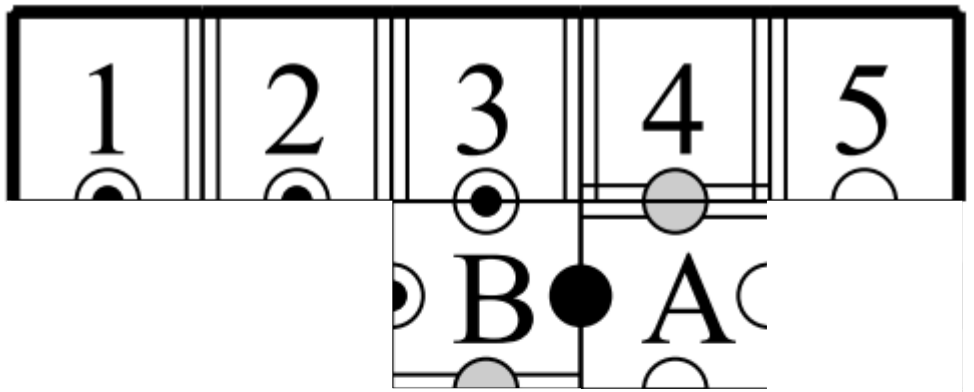

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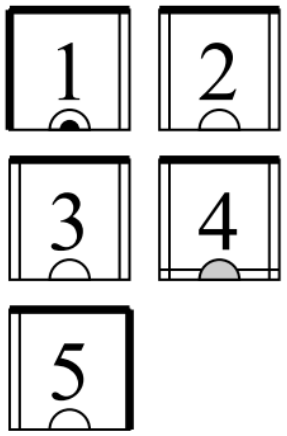

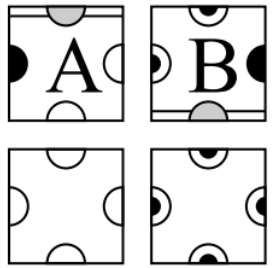

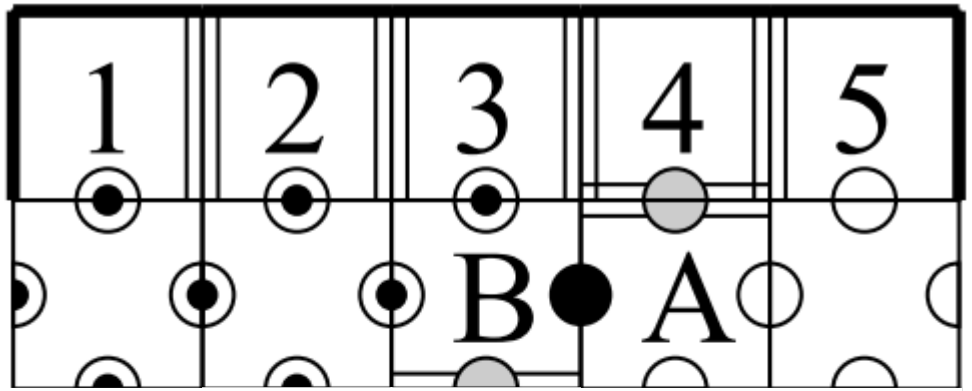

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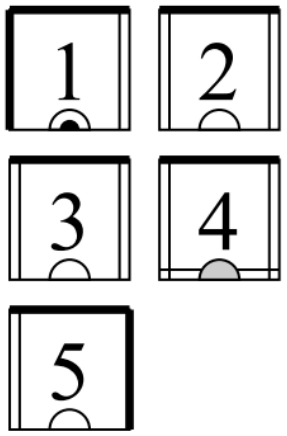

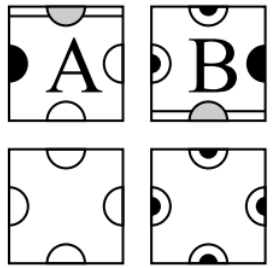

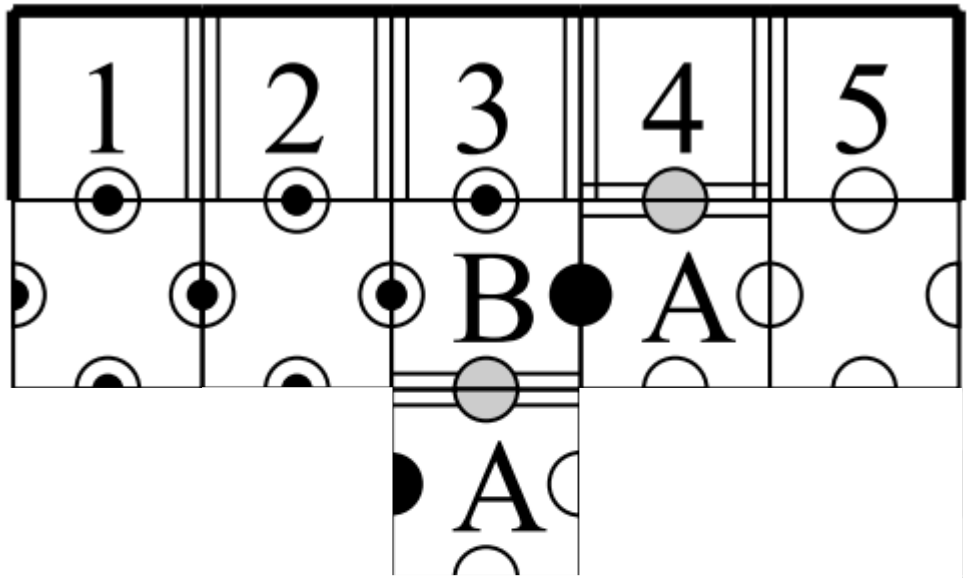

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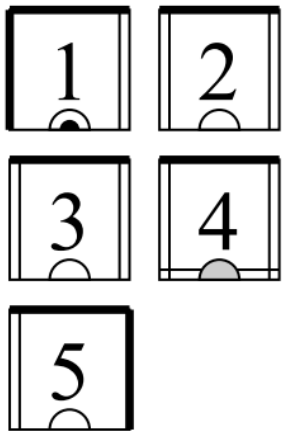

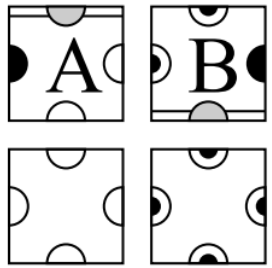

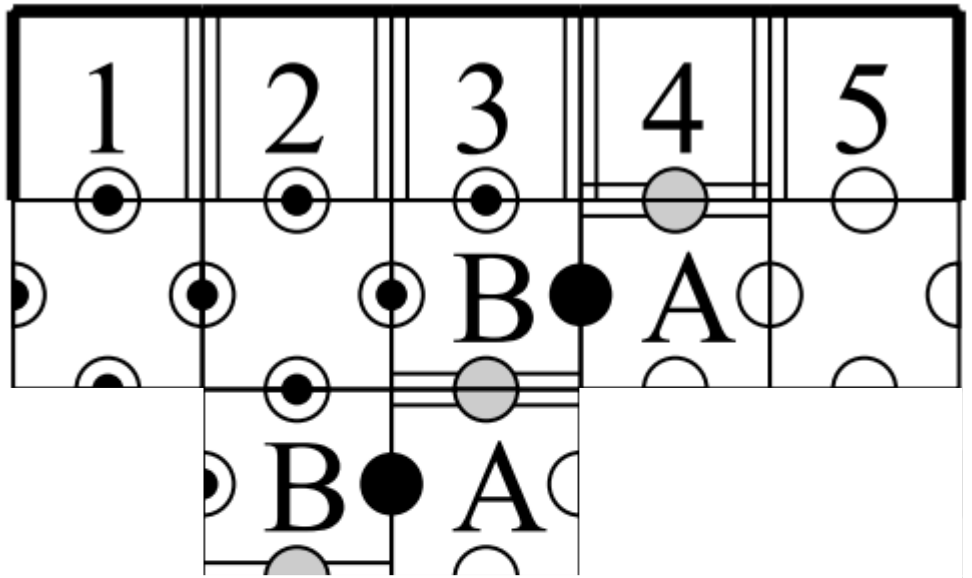

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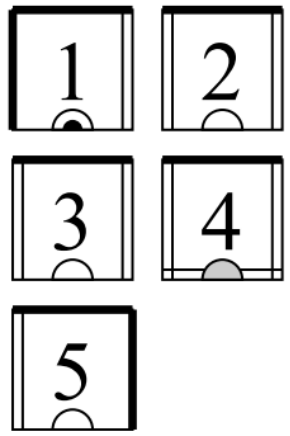

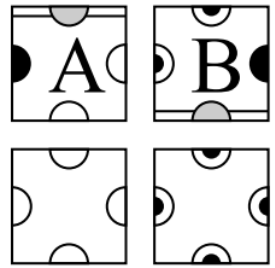

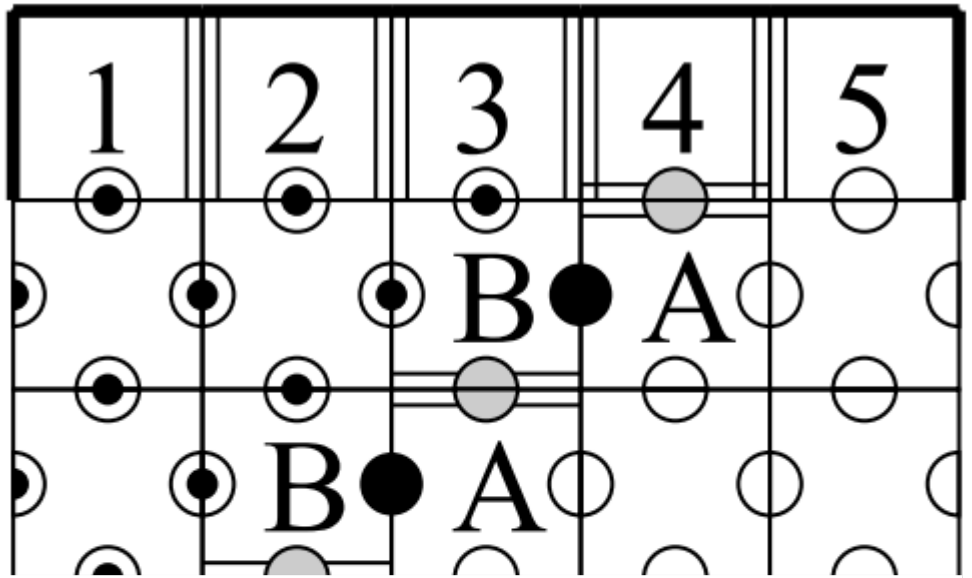

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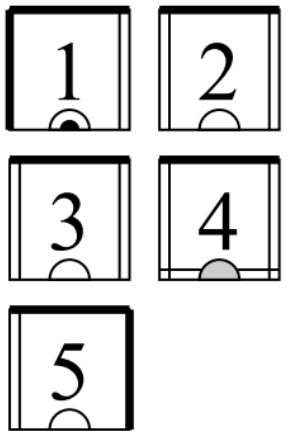

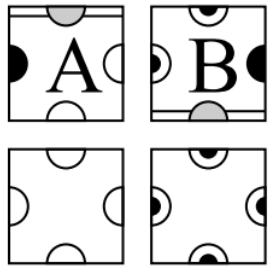

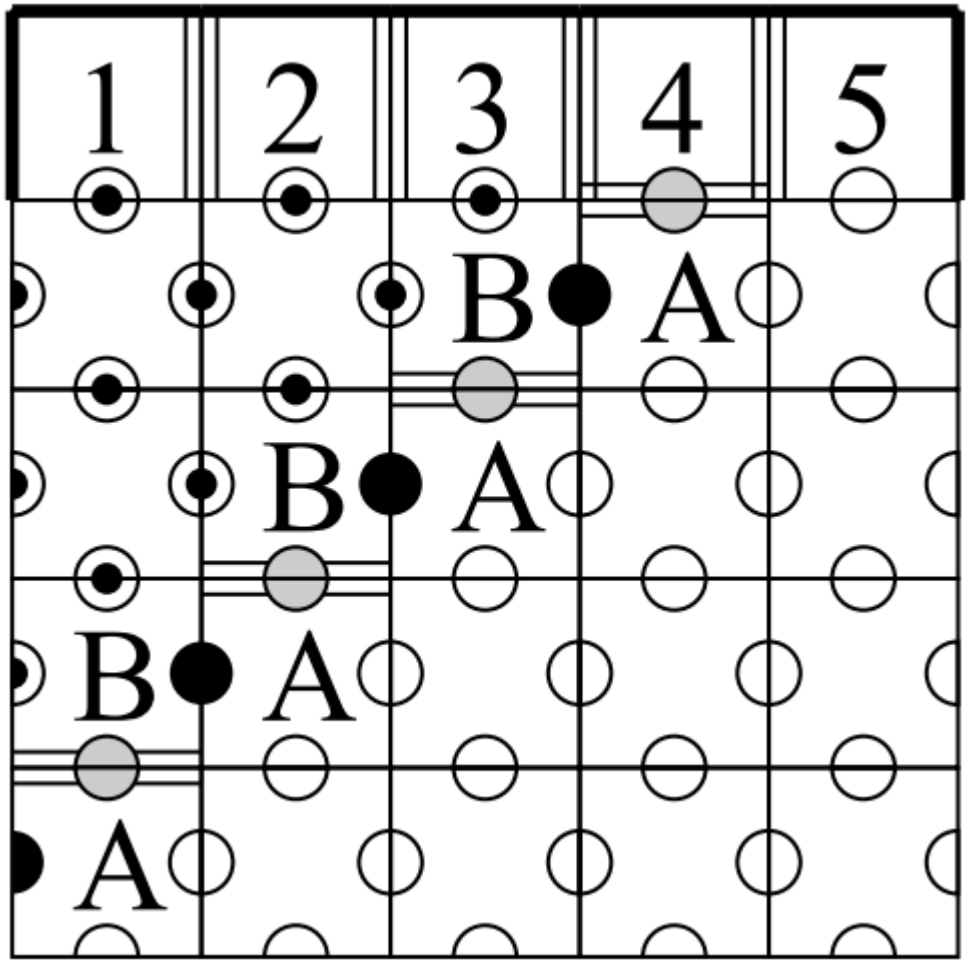

Tile complexity at temperature $\tau = 2$

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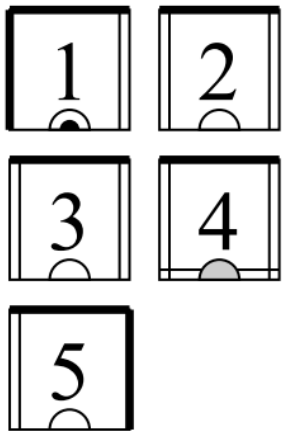
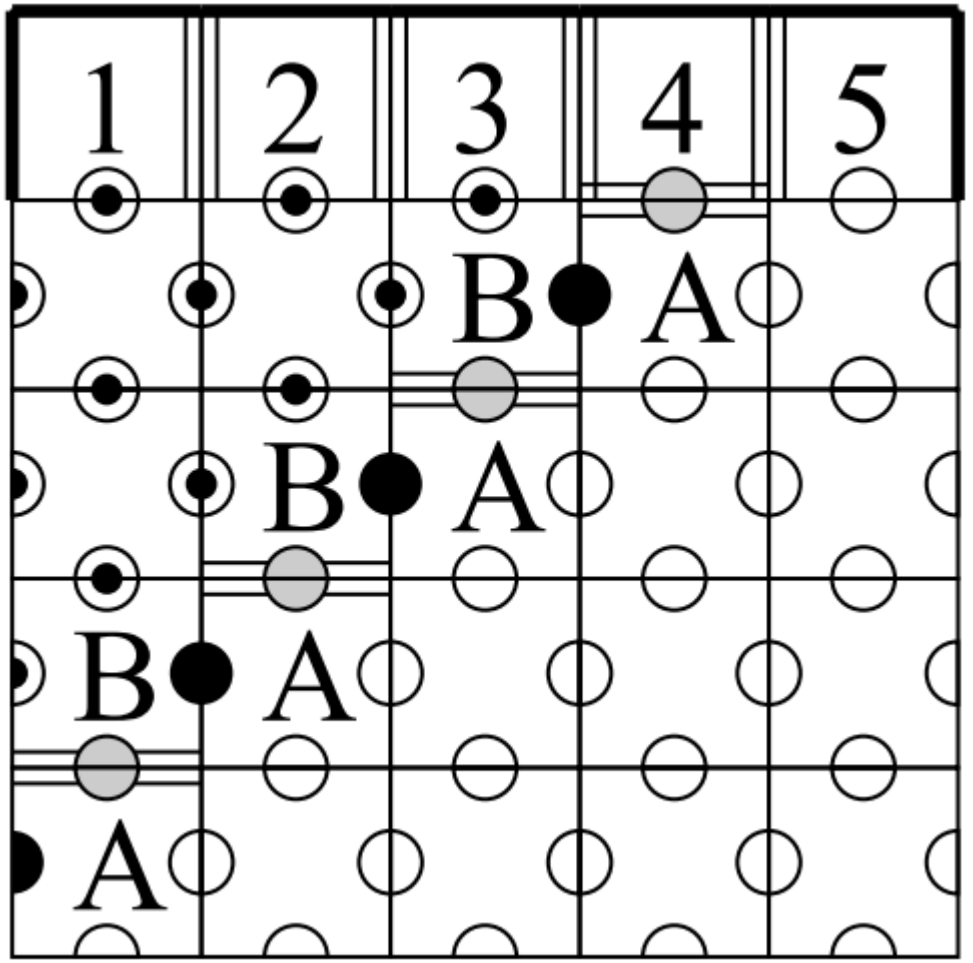
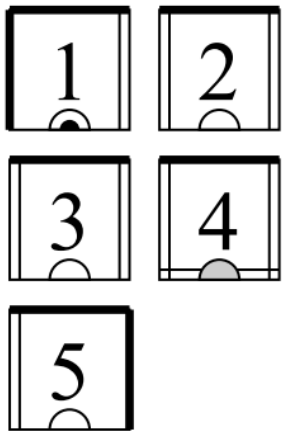


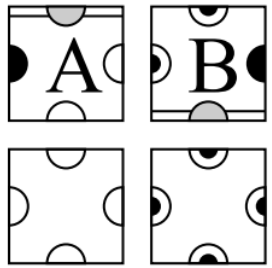


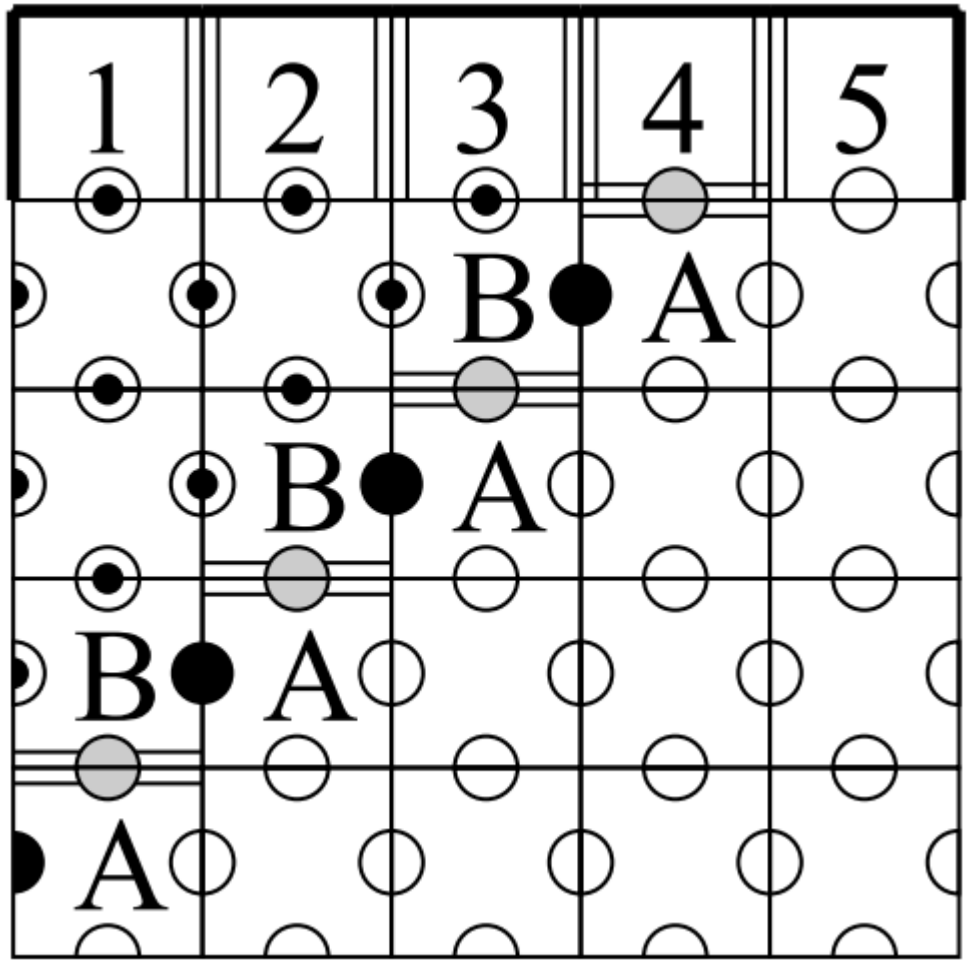
Tile complexity = *n* + 4

Tile complexity at temperature $\tau = 2$

Goal: complete a 1 x *n* line into an *n* x *n* square







Tile complexity = *n* + 4

How to get *sublinear* tile complexity?

Logarithmic tile complexity at temperature $\tau = 2$

Goal: rectangle of height n using $O(\log n)$ tile types

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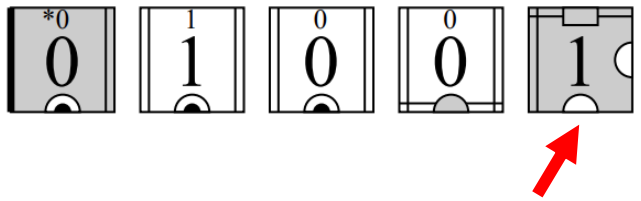

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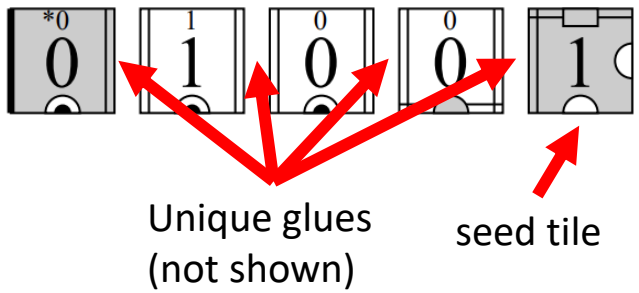

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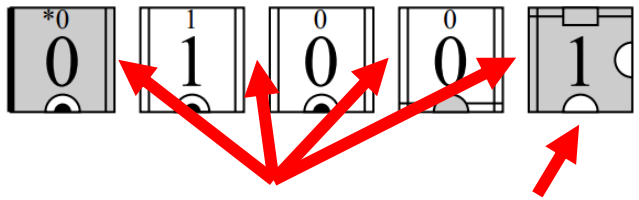
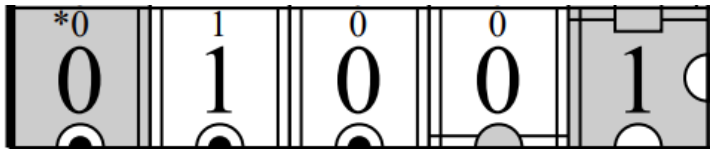


Unique glues (not shown)

seed tile

row encoding (a number related to) n



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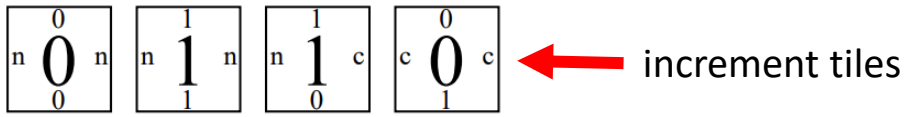
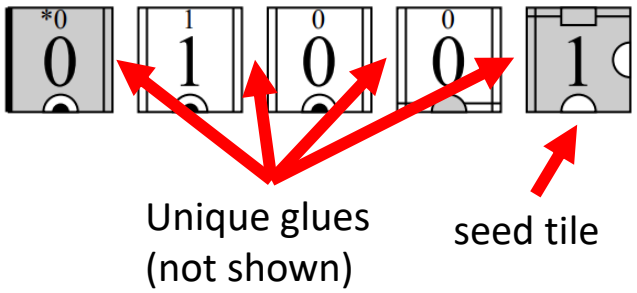
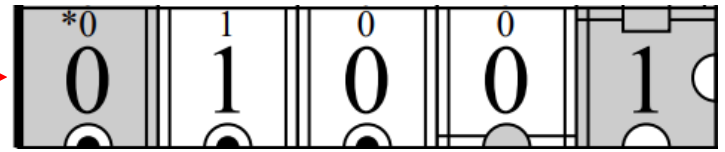




row encoding (a number related to) n →



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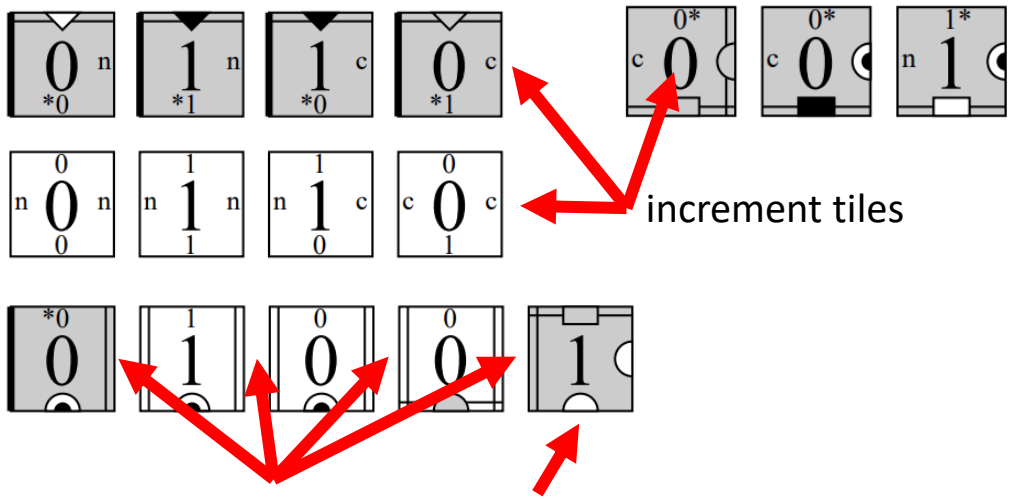

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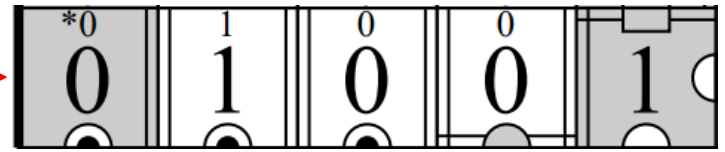

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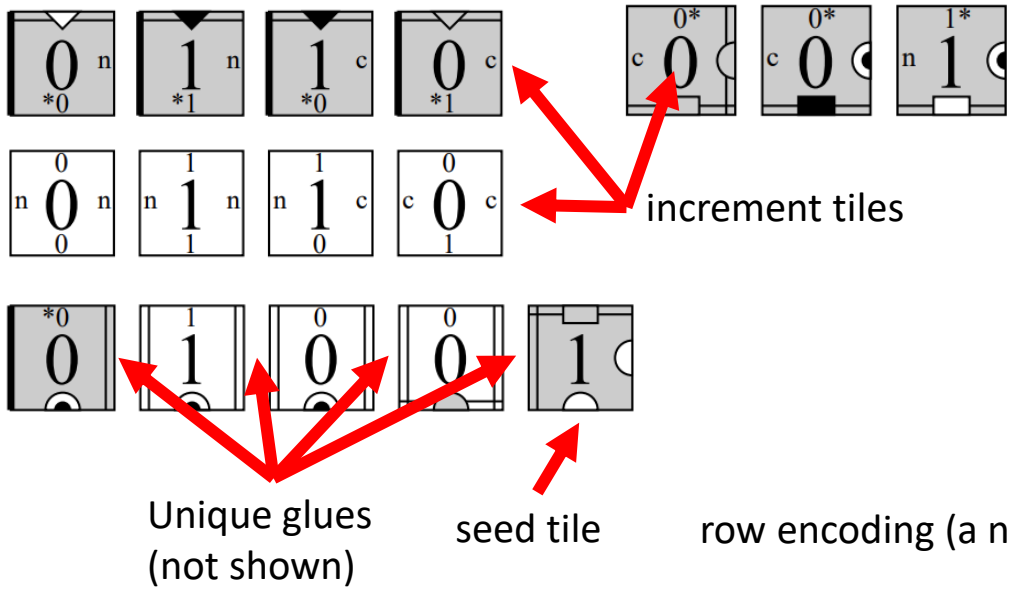

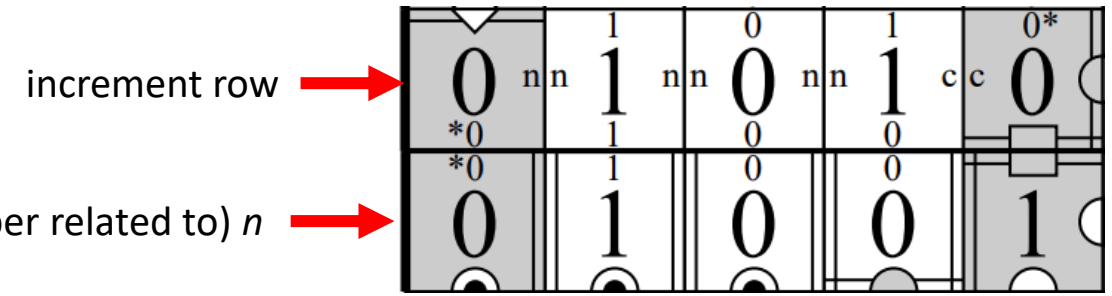

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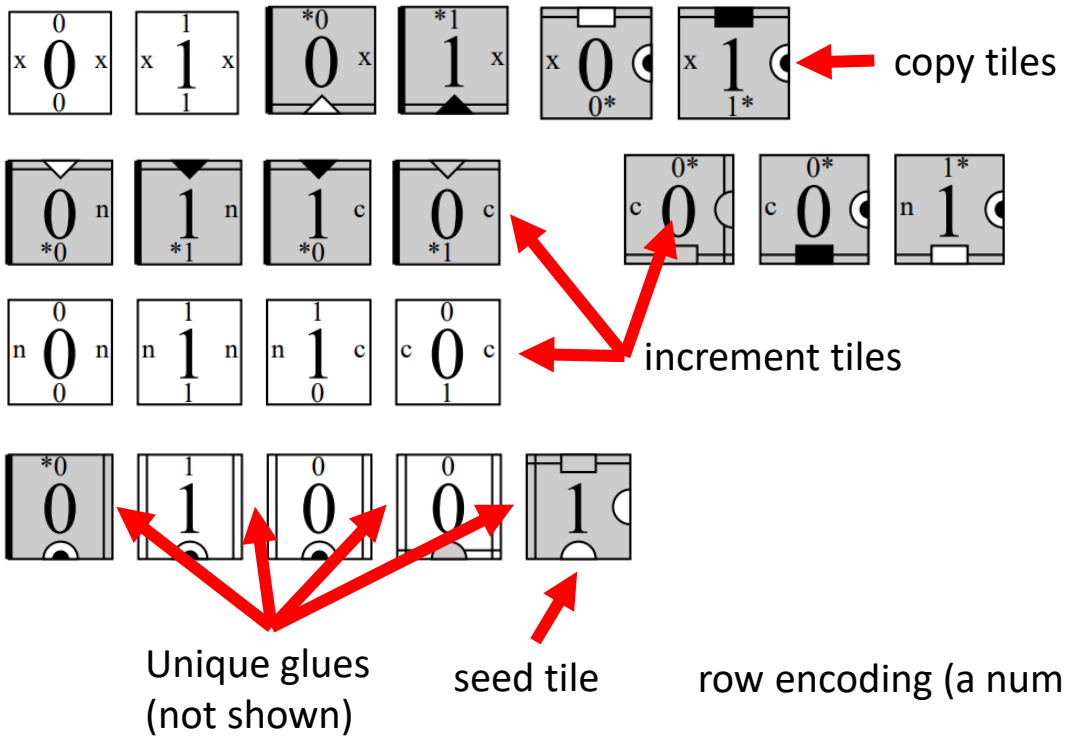

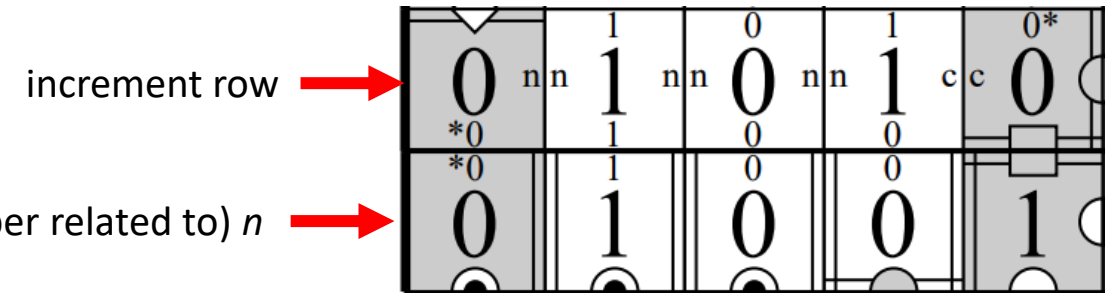

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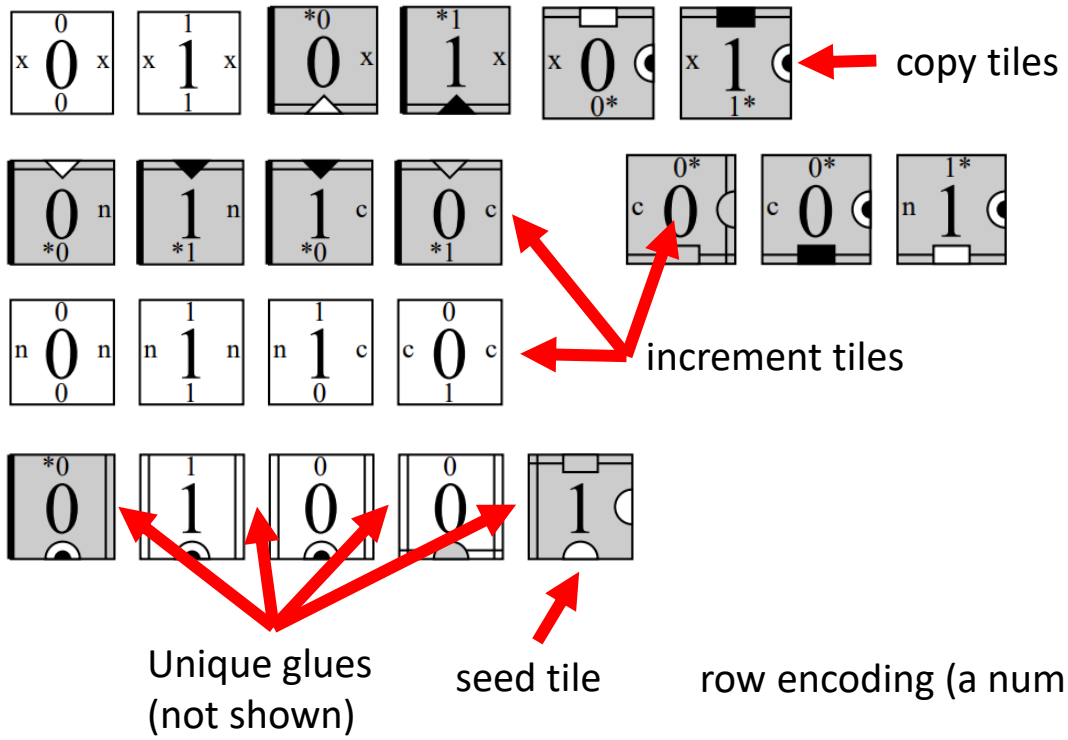

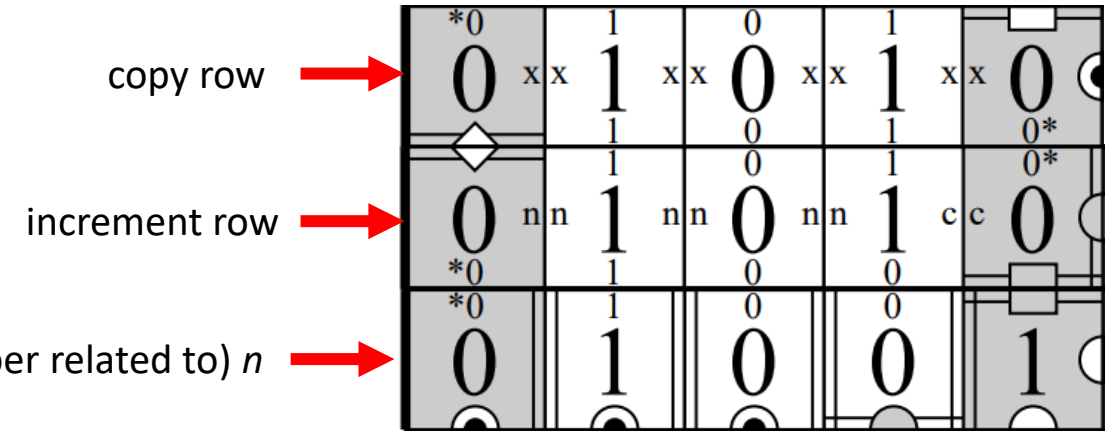

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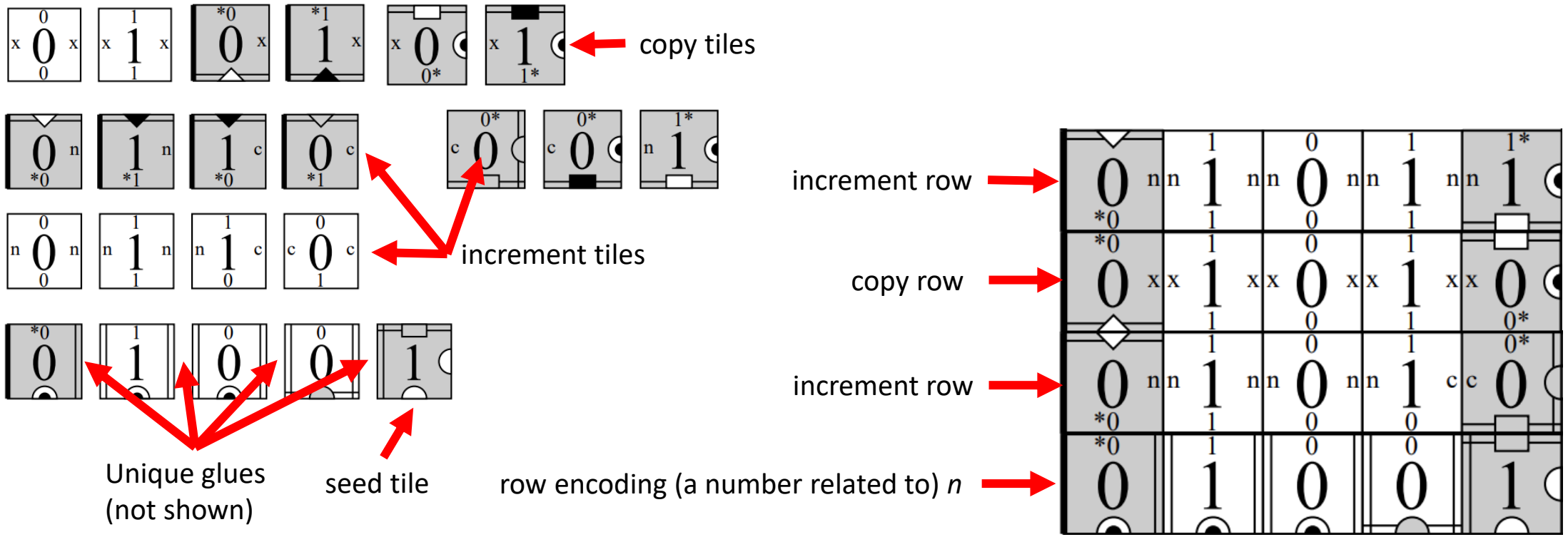

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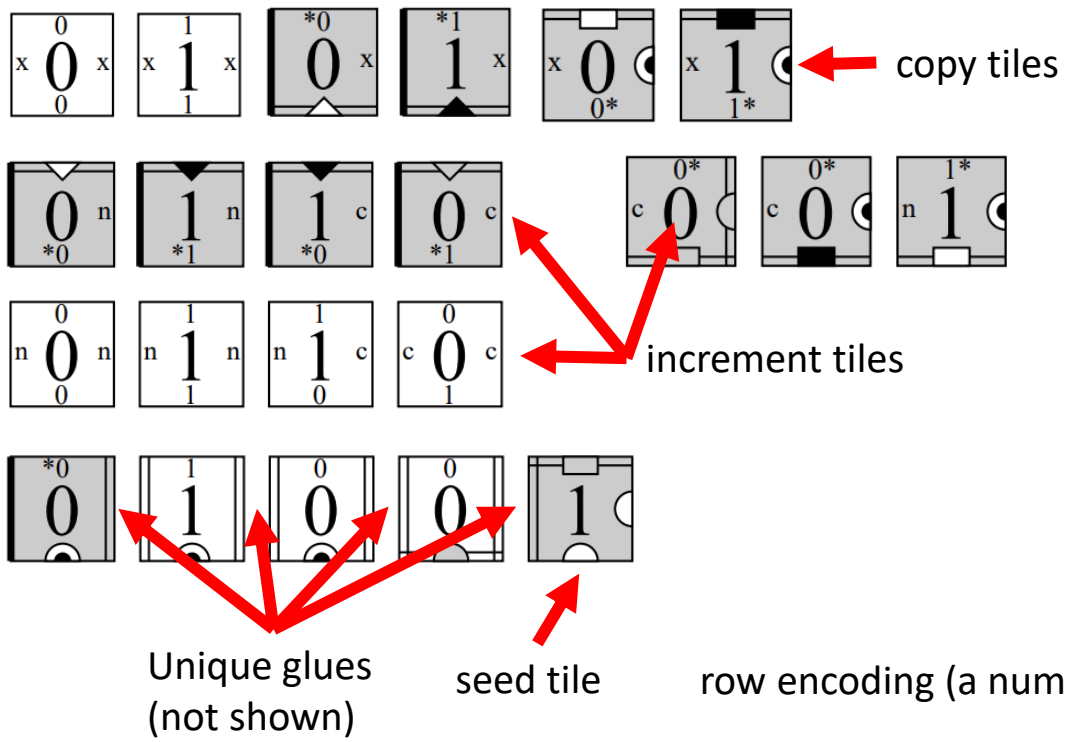
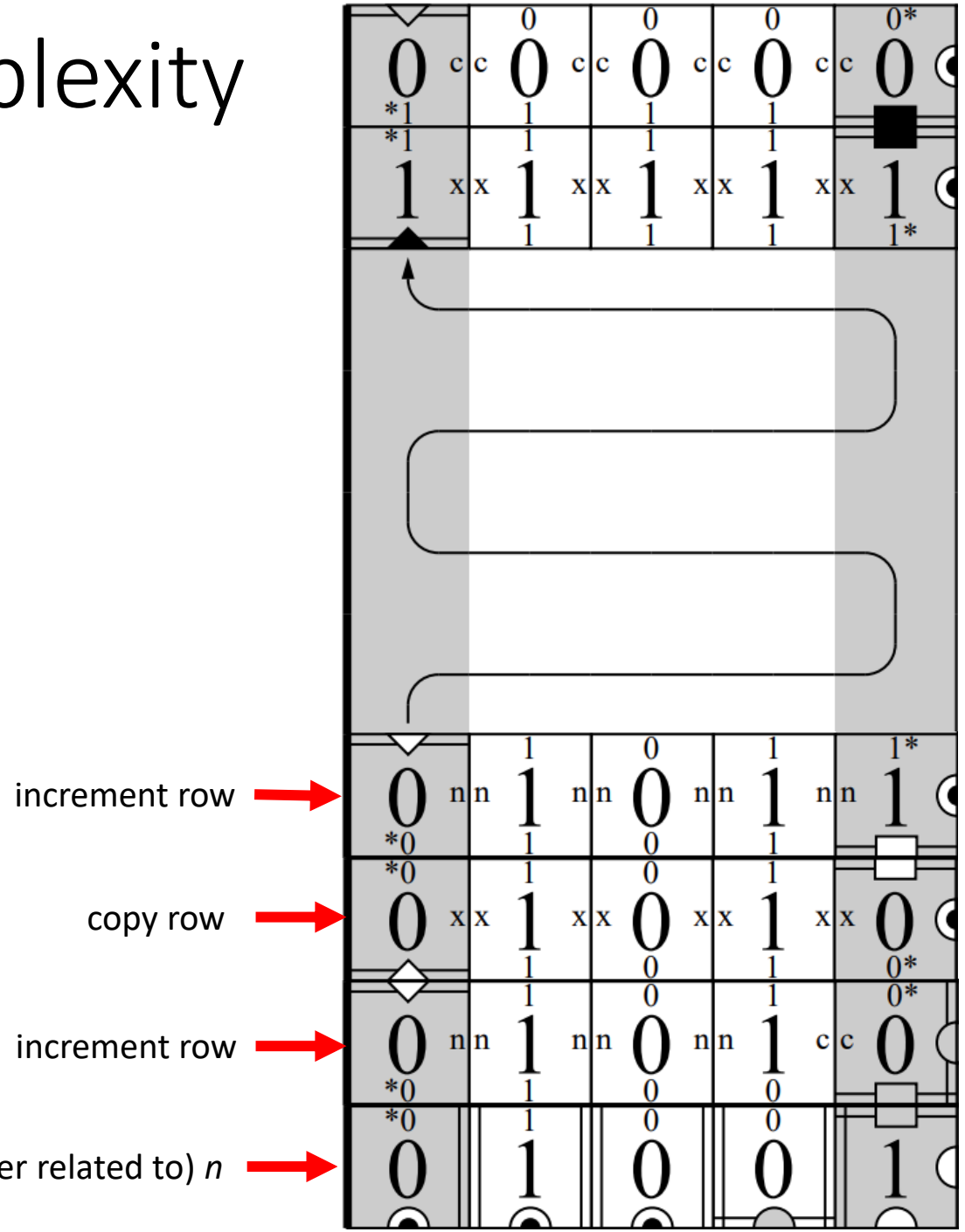
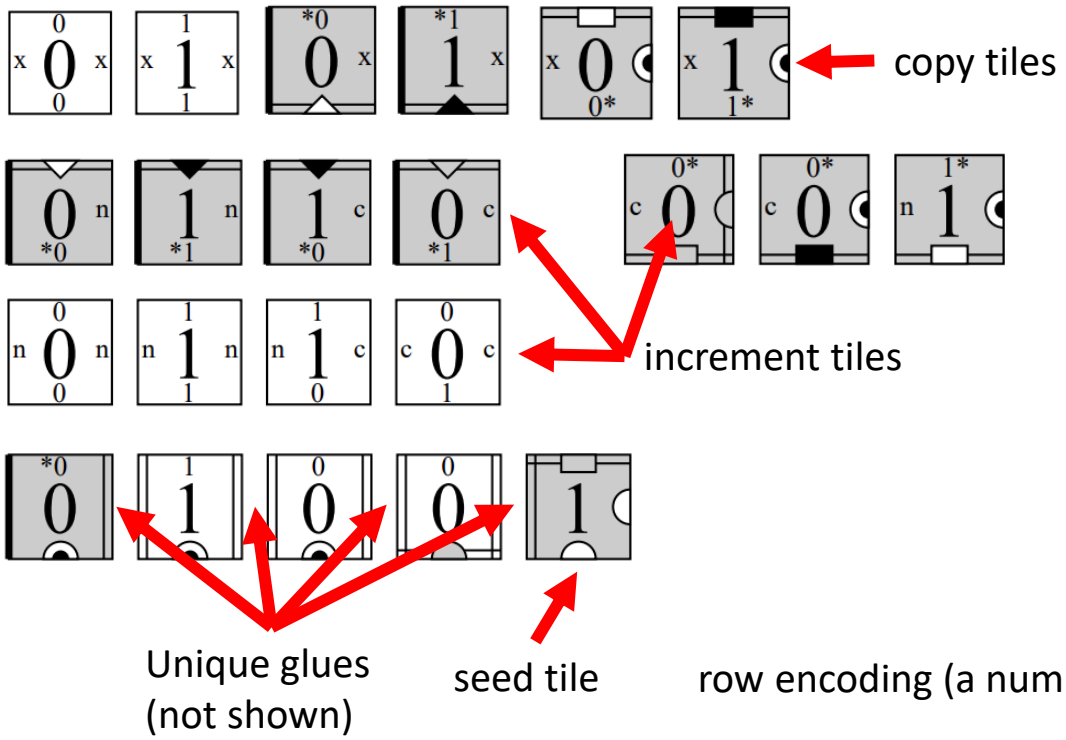


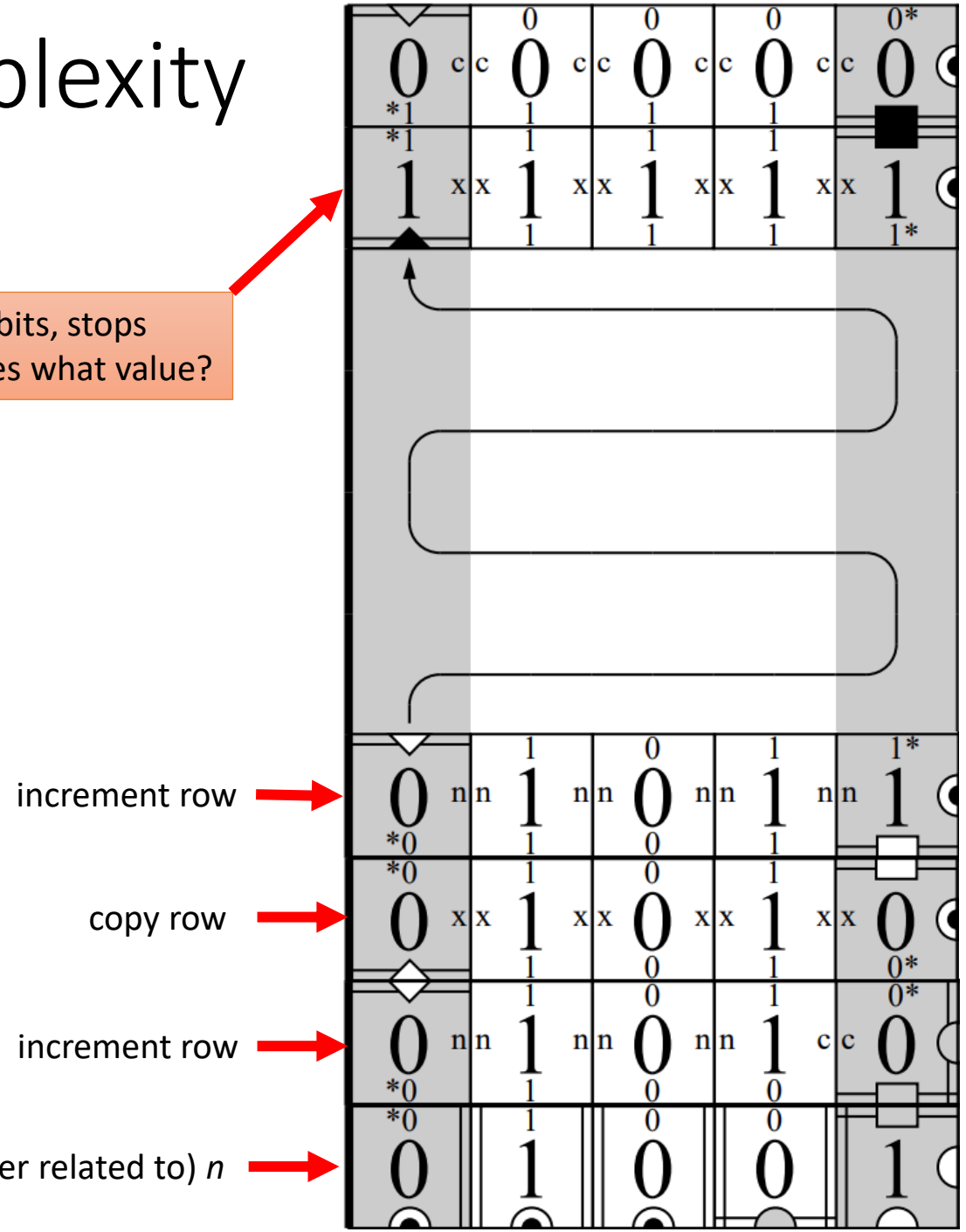


Logarithmic tile complexity at temperature $\tau = 2$

Goal: rectangle of height n using $O(\log n)$ tile types

for width of k bits, stops when it reaches what value?





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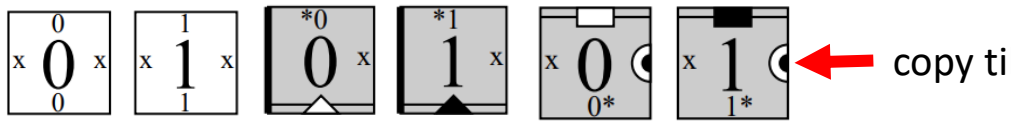
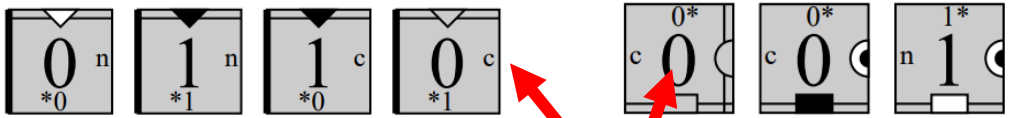
"zig-zag counter"



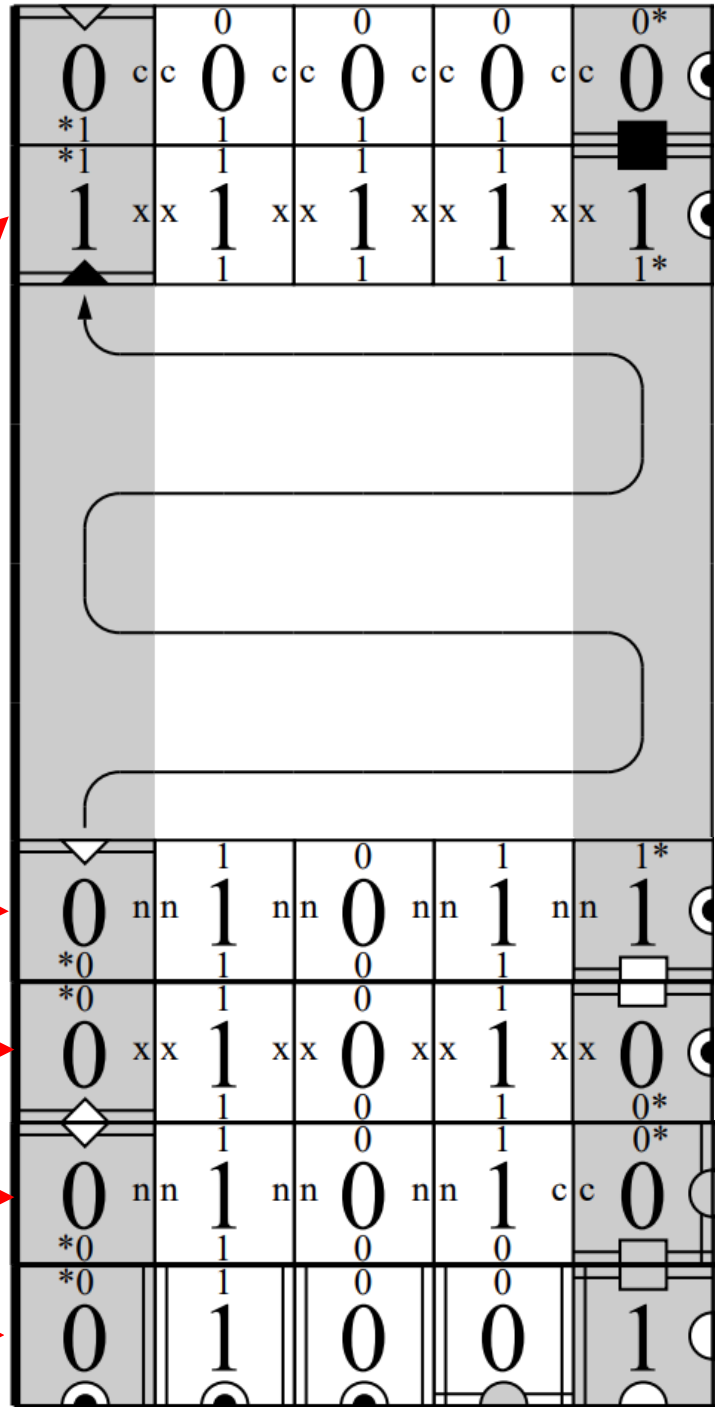
copy tiles



increment tiles

Unique glues (not shown)

seed tile



increment row

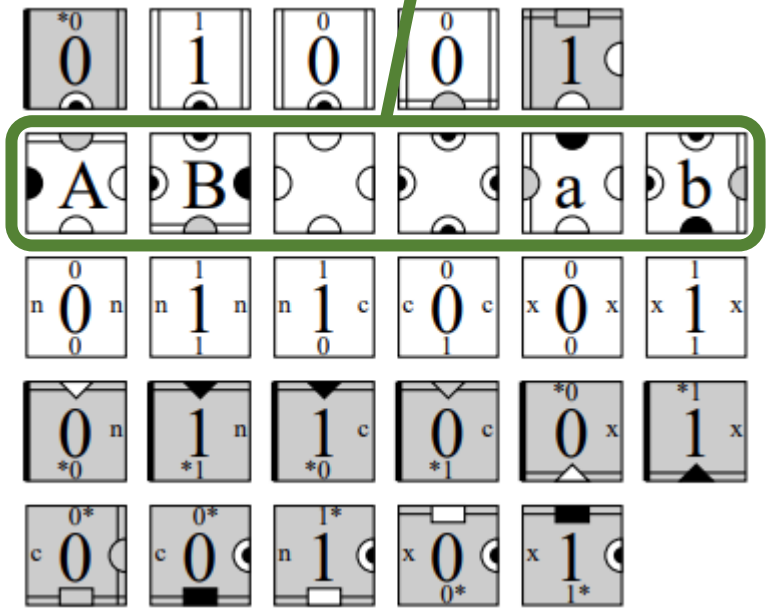
copy row

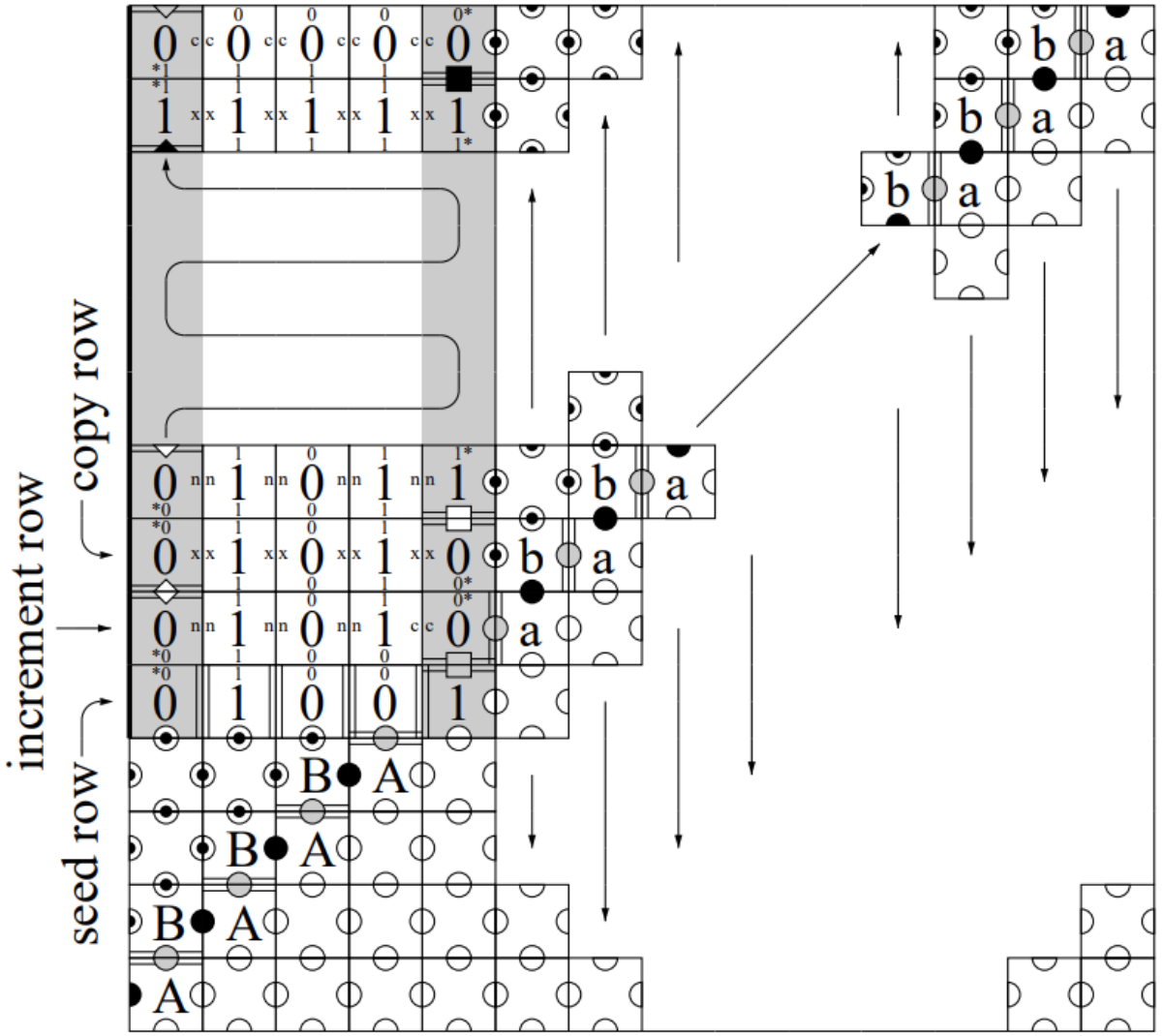
increment row

row encoding (a number related to) n

Logarithmic tile complexity at temperature $\tau = 2$

A few more “filler” tiles complete the ≈n x log n rectangle into an n x n square.

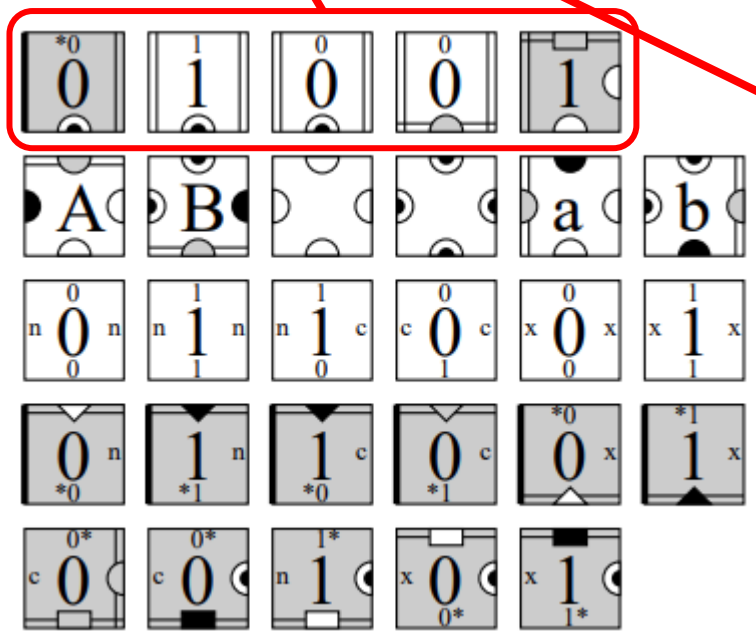


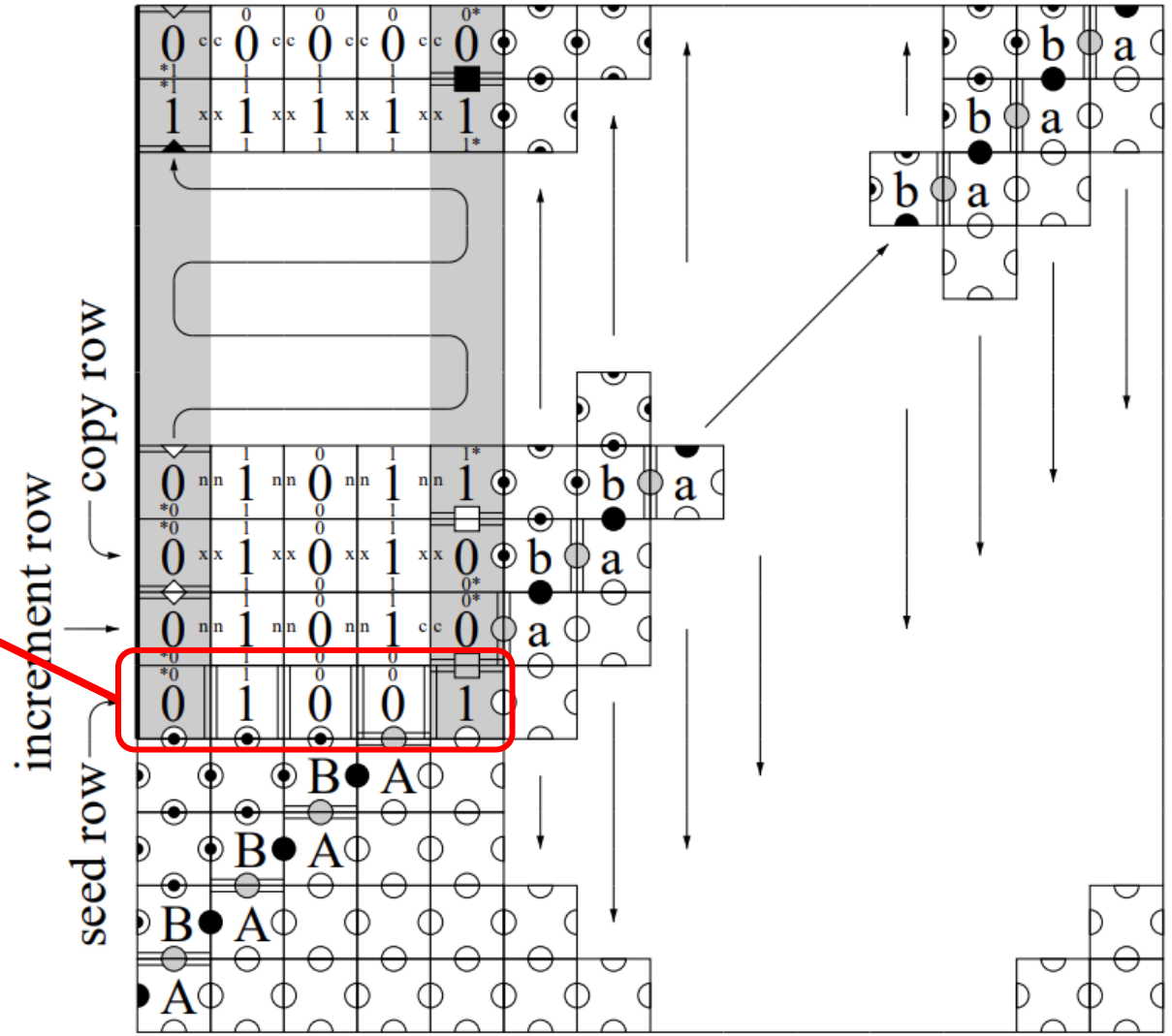


Logarithmic tile complexity at temperature $\tau = 2$

A few more "filler" tiles complete the $\approx n \times \log n$ rectangle into an $n \times n$ square.

tile complexity = $\log n + 23$





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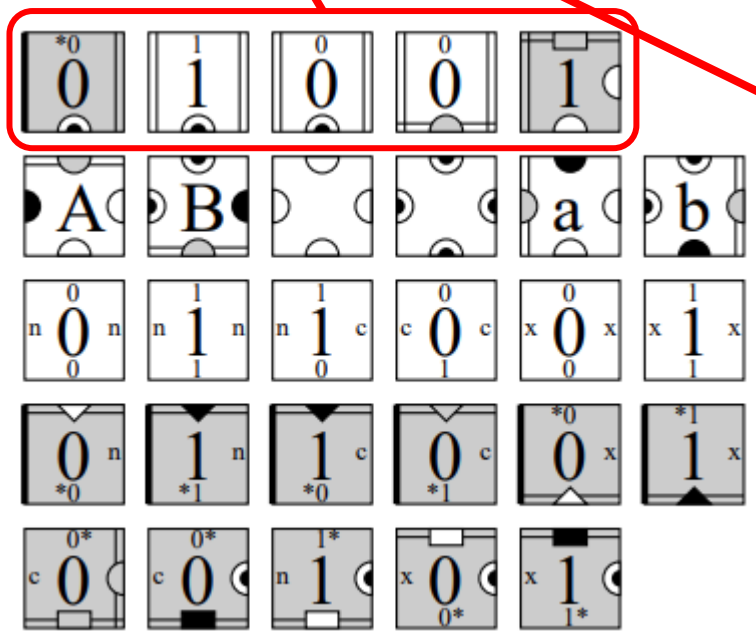

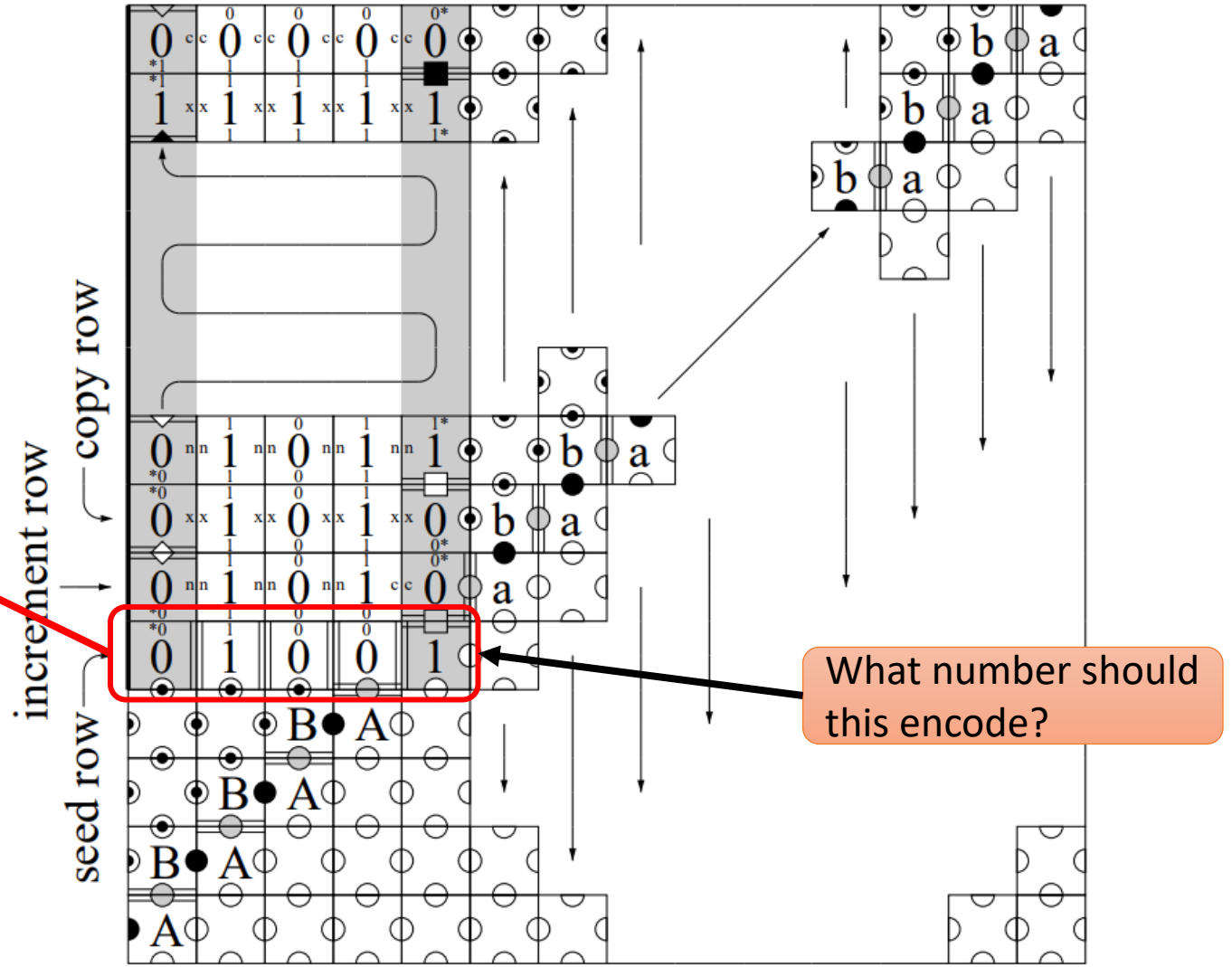

$\Omega(\log n / \log \log n)$ tile complexity lower bound for n x n squares

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$\Omega(\log n / \log \log n)$ tile complexity lower bound for n x n squares

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    - We'll show that it's fewer than p.

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    - We'll show that it's fewer than p.
  - There are p squares with width n between $p+1$ and $2p$; each needs a different tile system.
  - By pigeonhole, some n x n square cannot be assembled with $< \frac{1}{4} \log p / \log \log p$ tile types.
  - Since $p \leq n/2$, we have $\frac{1}{4} \log p / \log \log p \leq \frac{1}{4} \log n / \log \log n$.

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  - Since $p \leq n/2$, we have $\frac{1}{4} \log p / \log \log p \leq \frac{1}{4} \log n / \log \log n$.
  - Since we can do this for every positive integer p, there are infinitely many n that require more than $\frac{1}{4} \log n / \log \log n$ tile types (a stronger result holds: "most" values of n require that many)

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- How many tile systems? $c \cdot d = k(4k)^{4k}$

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- Recall $k = \frac{1}{4} \log p / \log \log p$; by algebra (see notes), $k^2(4k)^{4k} < p$.

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- Number of tile systems with <u>exactly</u> k tile types: $\leq k(4k)^{4k}$
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- Recall $k = ¼ \log p / \log \log p$; by algebra (see notes), $k^2(4k)^{4k} < p$.
- By pigeonhole principle, for some width n with $p < n \leq 2p$, the n x n square is not self-assembled by one of these $k^2(4k)^{4k}$ tile systems. Since those are all the tile systems with at most k tile types, the n x n square requires **more** than $¼ \log p / \log \log p$ tile types to self-assemble. **QED**

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  - https://en.wikipedia.org/wiki/Kolmogorov_complexity

Fact: “most” integers *n* require ≥ log *n* bits to “describe”.
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Note: we're ignoring glue strengths here; adds 2 bits per glue to describe at temperature 2. (since there are 3 possible strengths 0, 1, 2);
see http://doi.org/10.1007/s00453-014-9879-3 for handling higher-temperature systems.

Which bound is tight?

1. All n x n squares can be assembled with $O(\log n)$ tile types; can we get it down to $O(\log n / \log \log n)$?
2. Or do we need $\Omega(\log n)$ tile types to assemble infinitely many n x n squares?

Improved upper bound: self-assembling an n x n square with $O(\log n / \log \log n)$ tile types

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Recall:

tile complexity =
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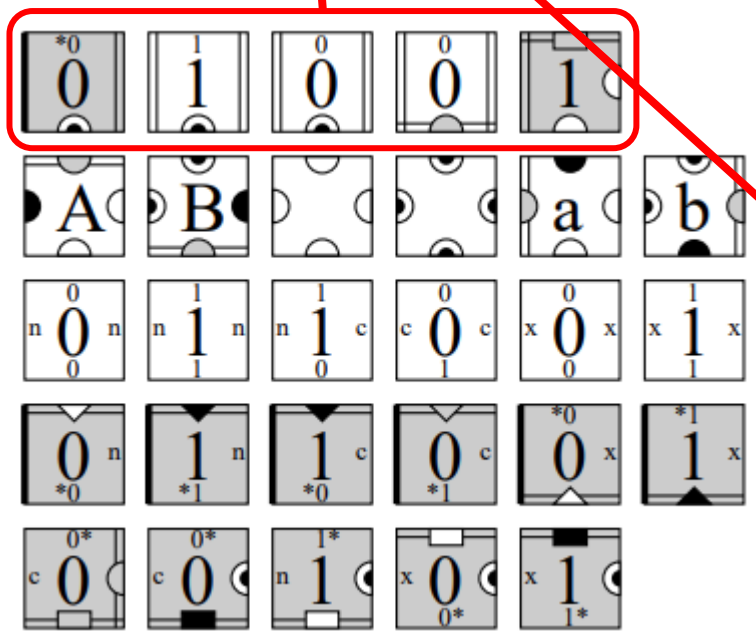

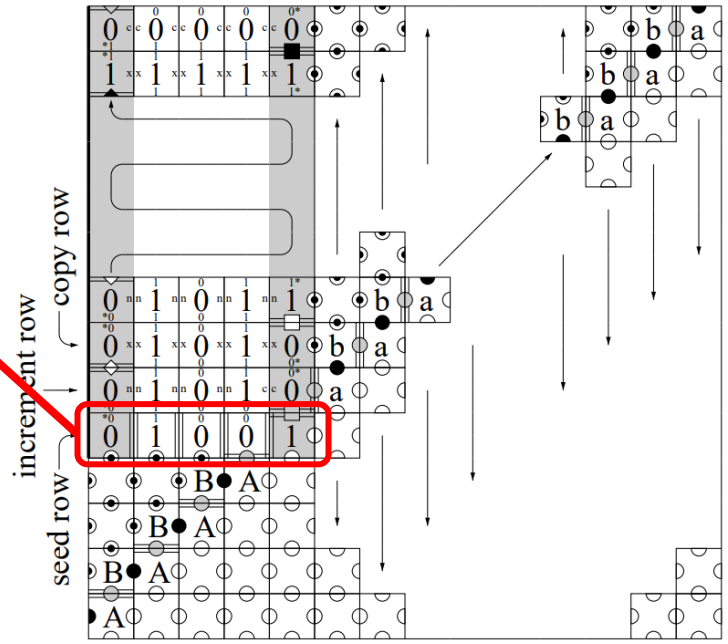

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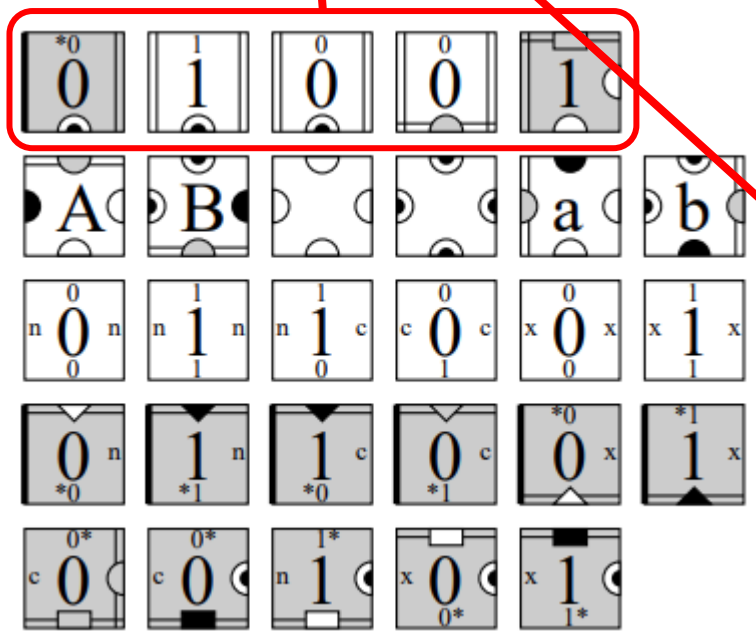

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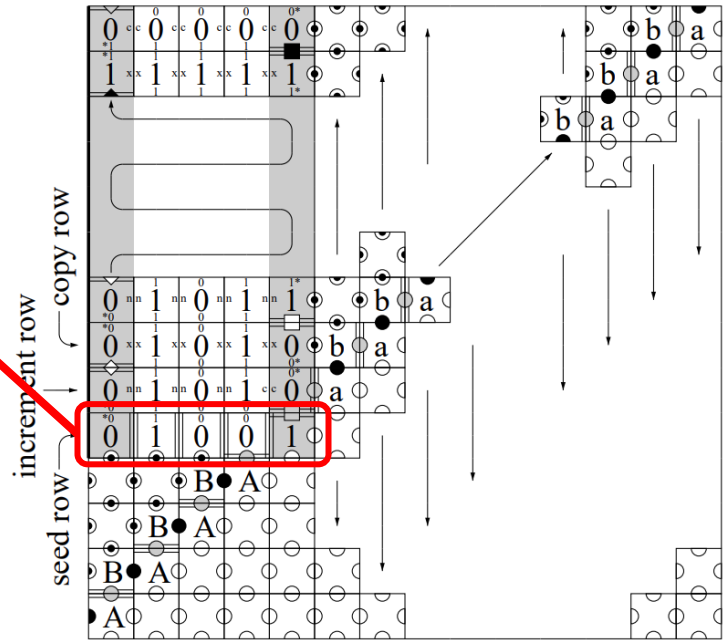

Idea:

1) Use same 23 tiles that turn the seed row encoding a binary integer n' (related to n) into an n x n square.

2) Create the binary seed row from only $\log n / \log \log n$ tiles.

Creating a row of log n glues with arbitrary bit string $s \in \{0,1\}^{\log n}$ using O(log n / log log n) tile types

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- Key idea: choose larger power-of-two base $b = 2^k$, with $b \approx \log n / \log \log n$, and convert from base b to base 2.

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- Key idea: choose larger power-of-two base $b = 2^k$, with $b \approx \log n / \log \log n$, and convert from base b to base 2.
- How many base-b digits needed to represent a log(n)-bit integer?

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- Each base-b digit is k bits
  - e.g., if $b=2^3=8$, then 0=000 1=001 2=010 3=011 4=100 5=101 6=110 7=111
  - e.g., the octal number 7125_8 in binary is 1110010101012

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  - e.g., the octal number 7125_8 in binary is 1110010101012
  - need $\log(n) / k = \log(n) / \log (\log n / \log \log n) = \log(n) / (\log \log n - \log \log \log n) \approx \log(n) / \log \log n$ base-b digits.

Creating a row of log *n* glues with arbitrary bit string $s \in \{0,1\}^*$ using log *n* / log log *n* tile types (i.e., base conversion from *b* to 2)

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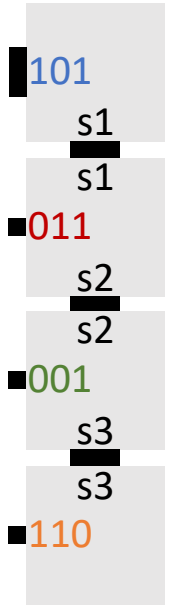

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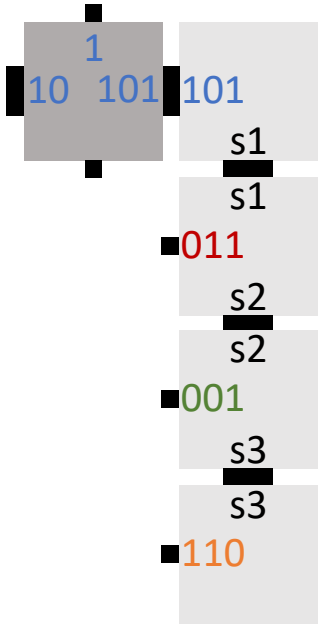

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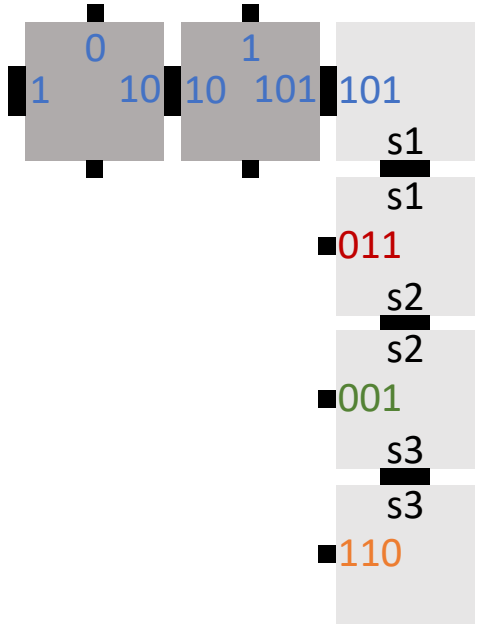

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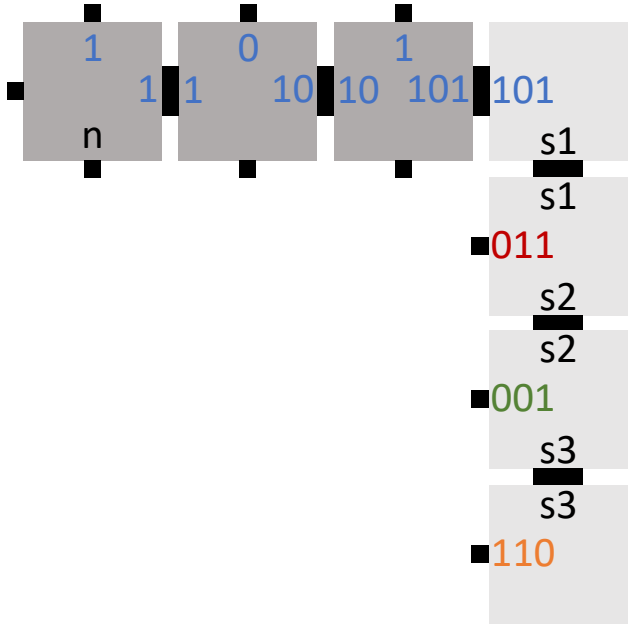

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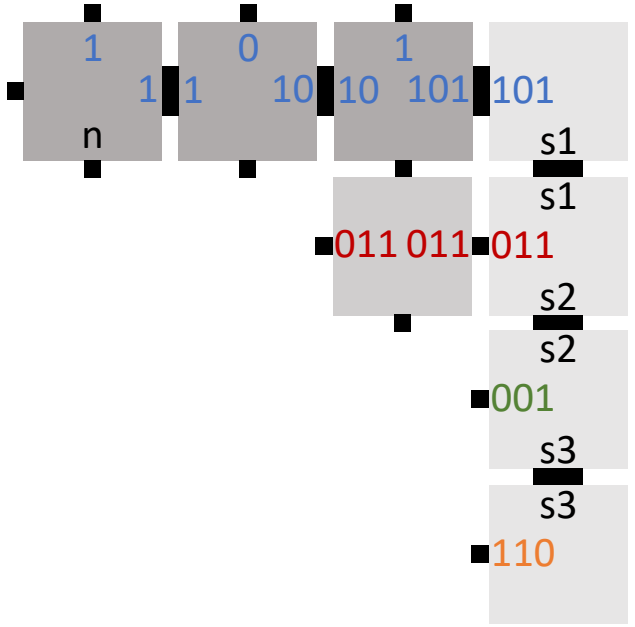

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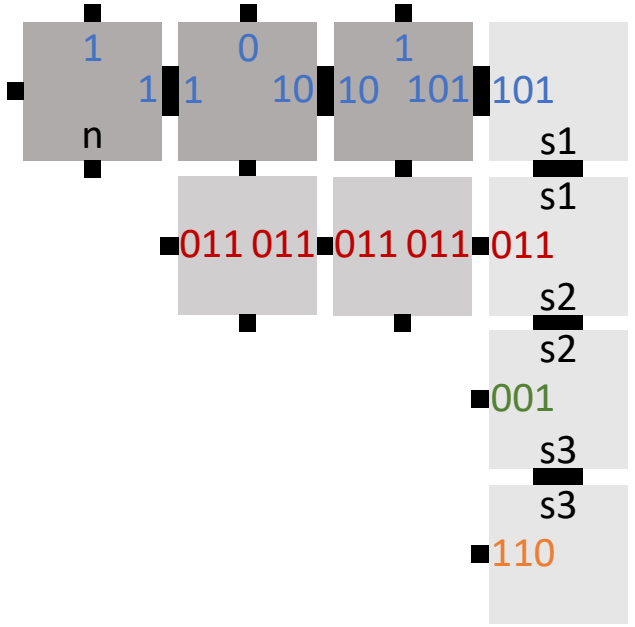

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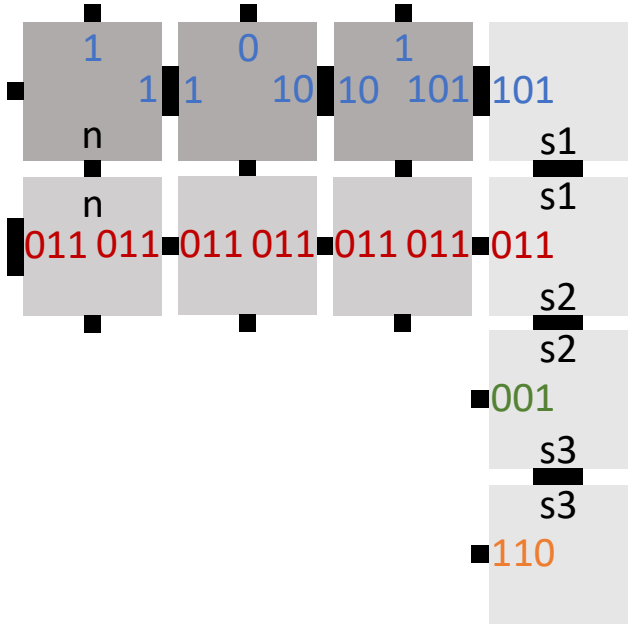

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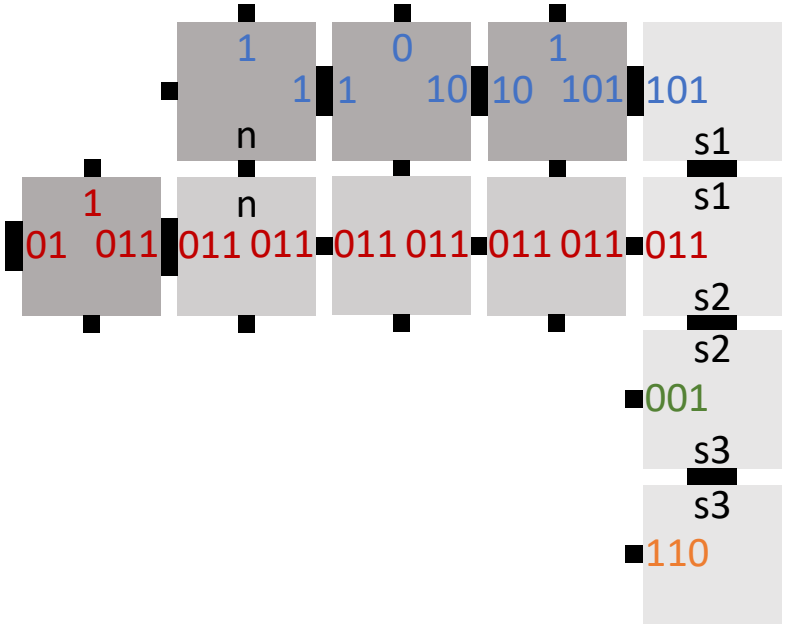

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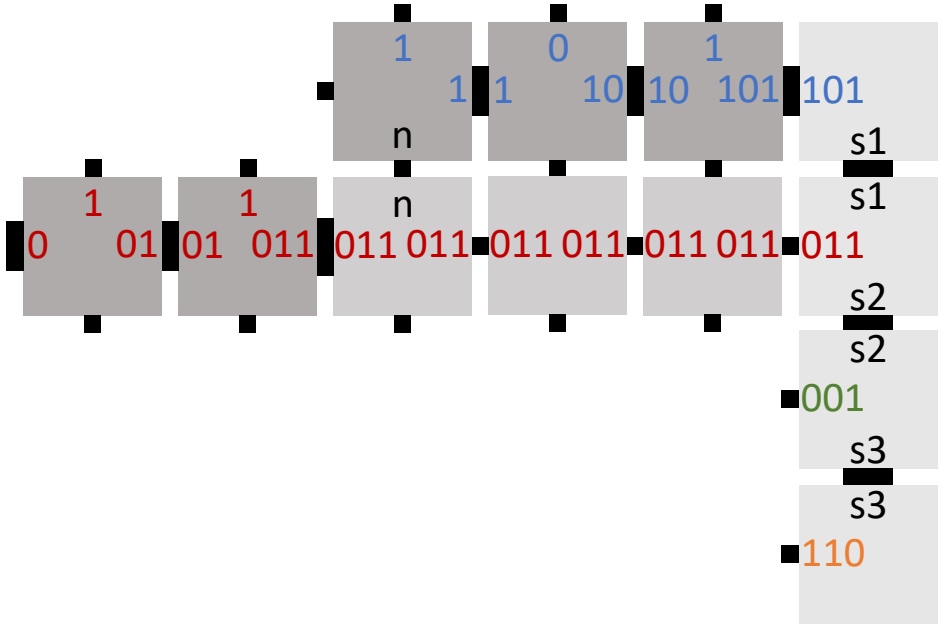

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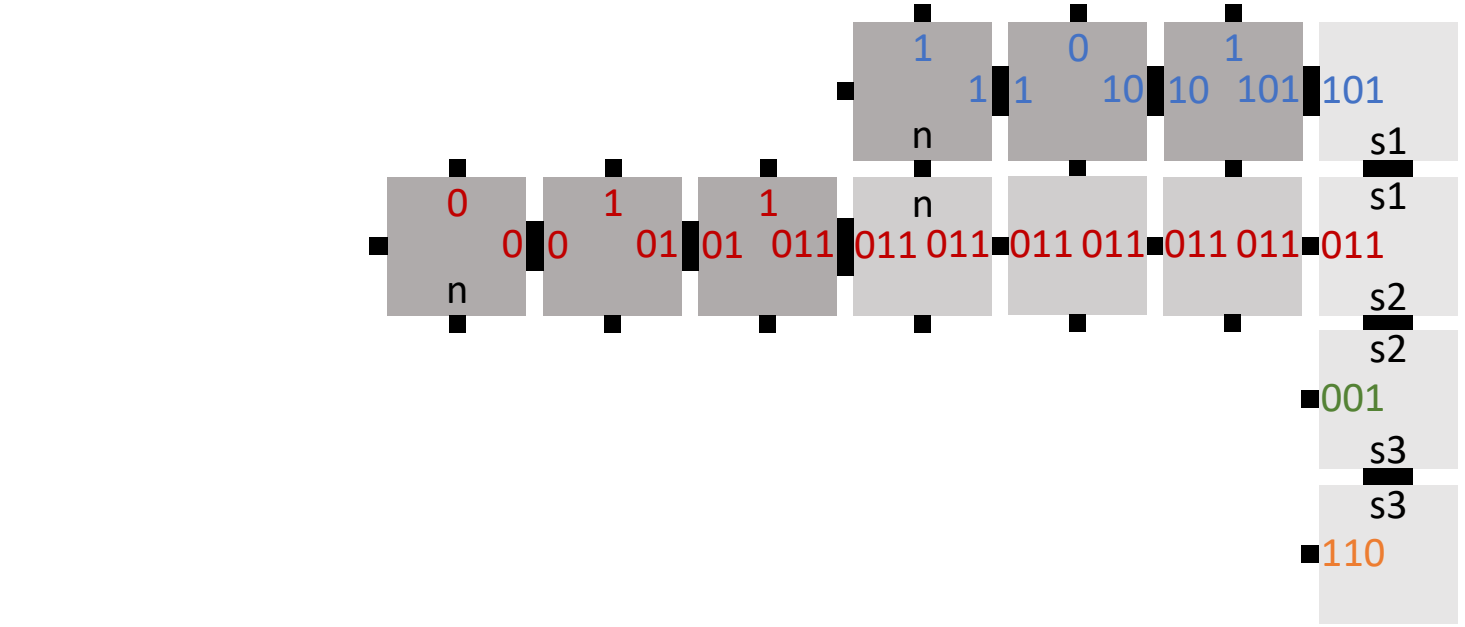

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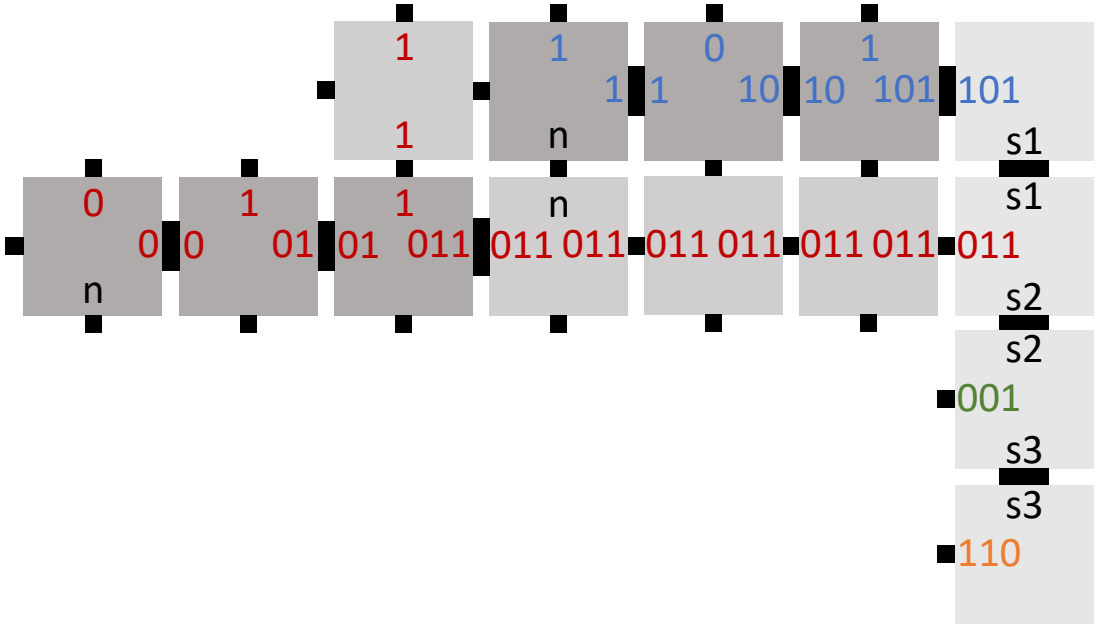

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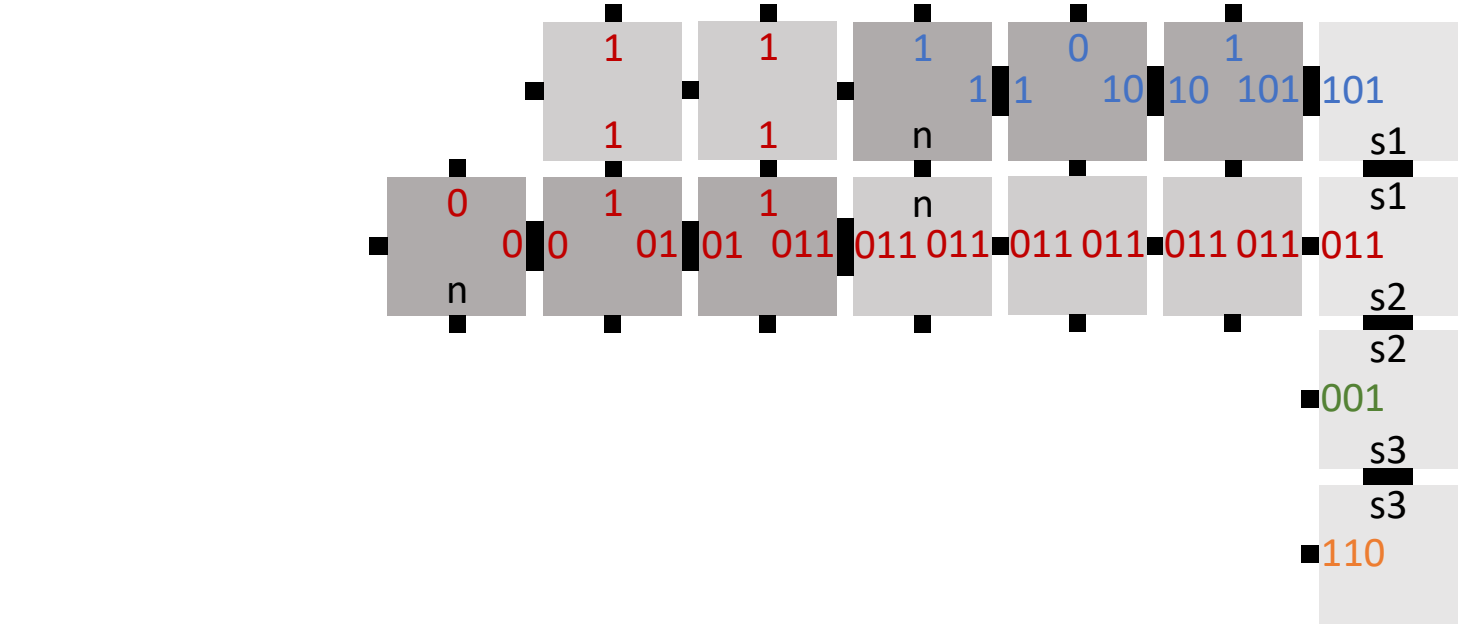

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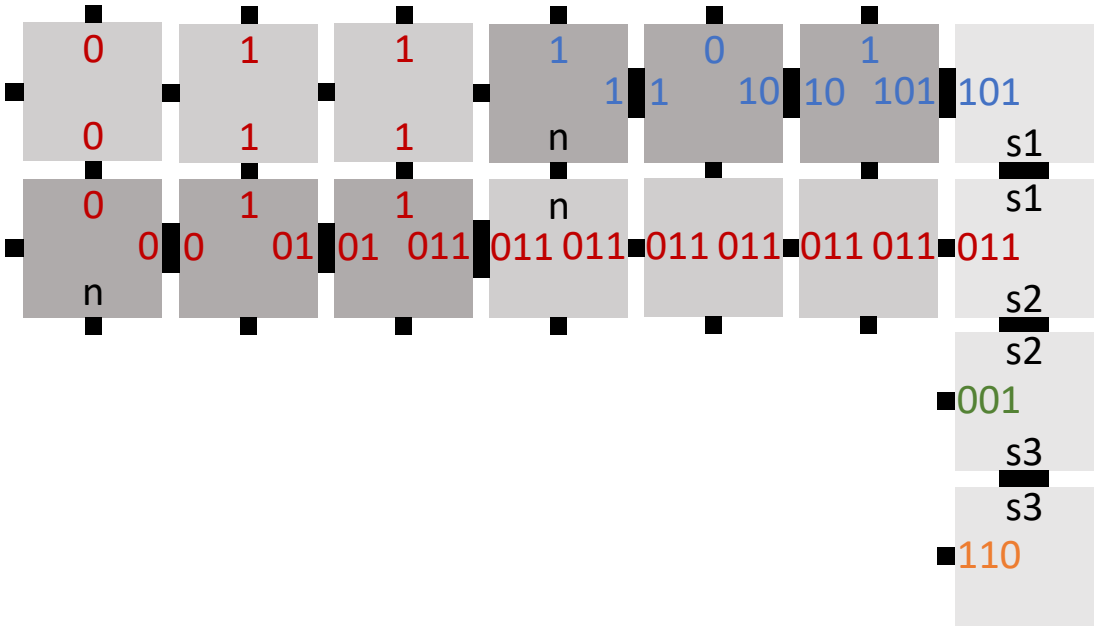

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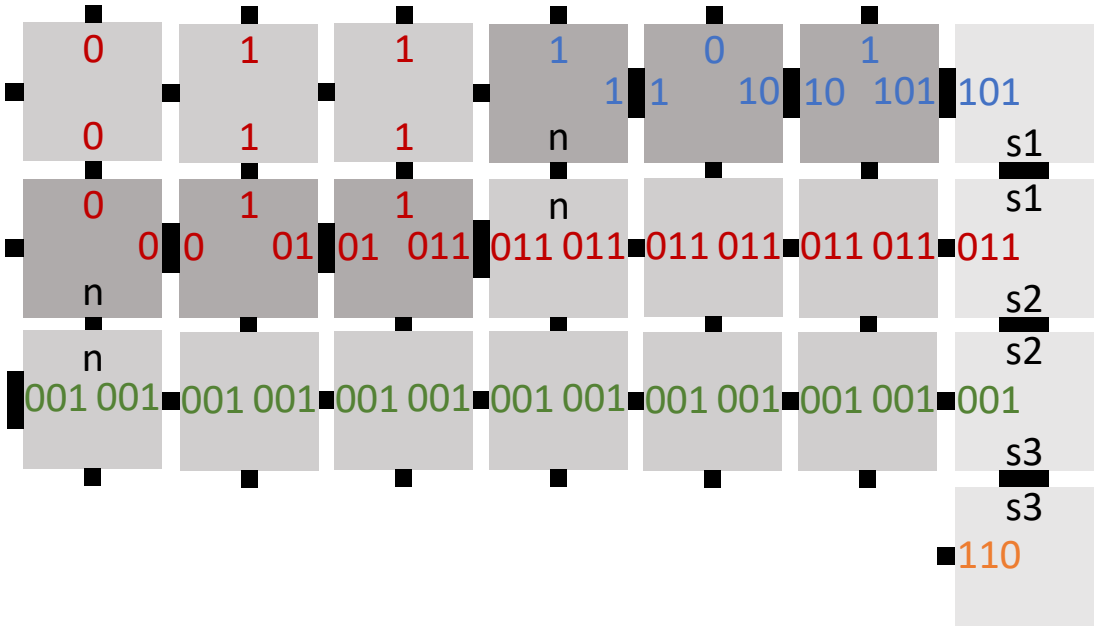

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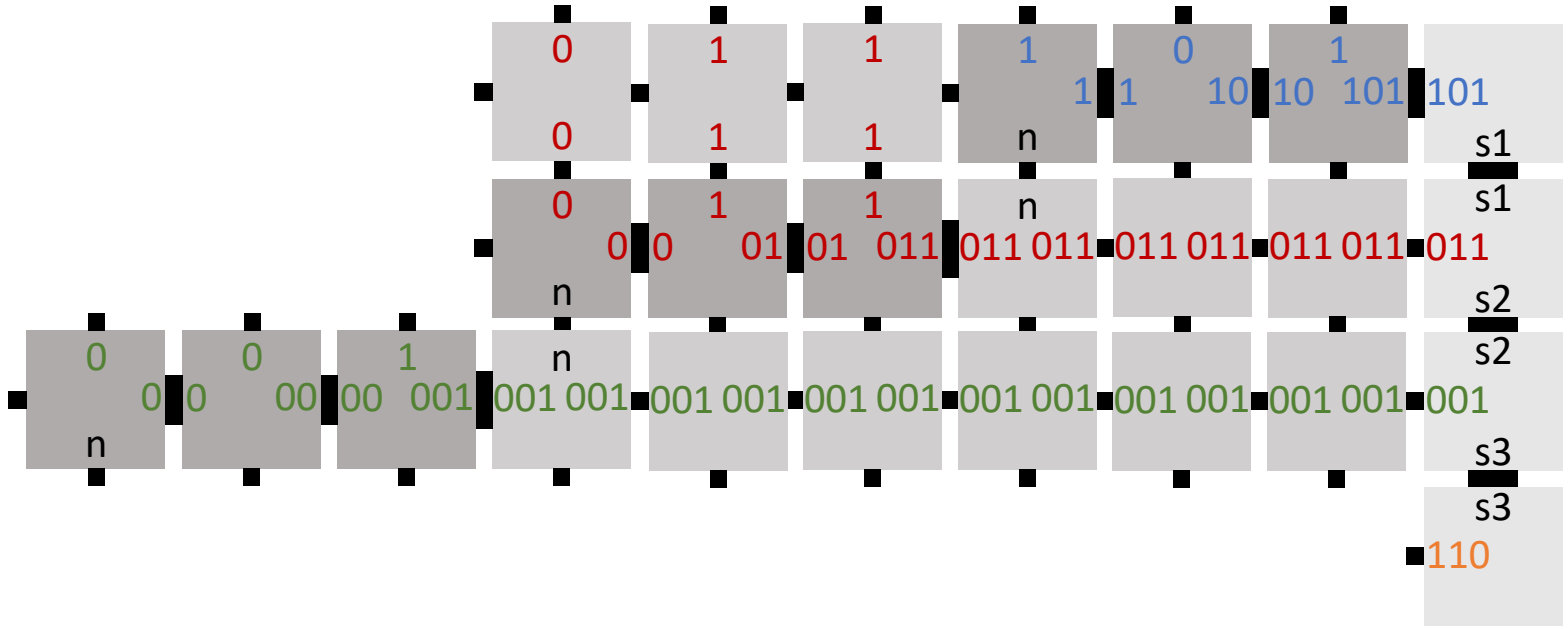

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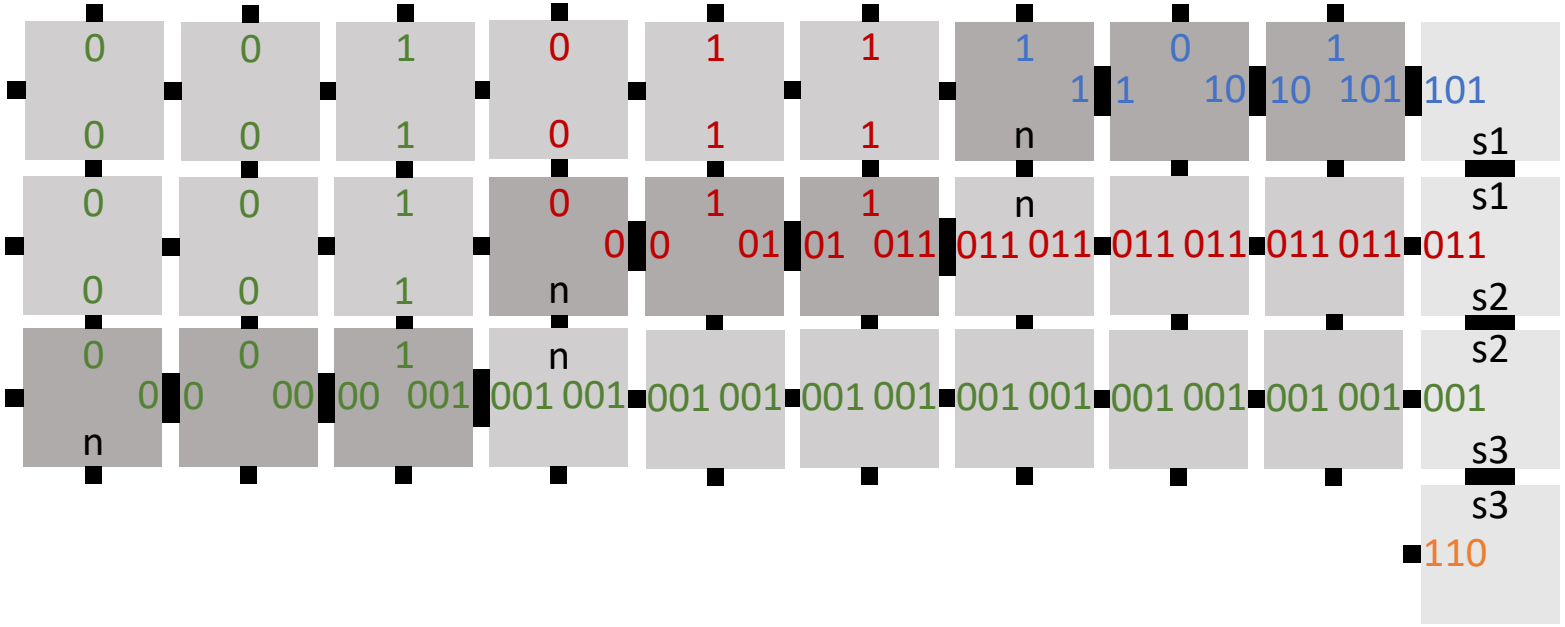

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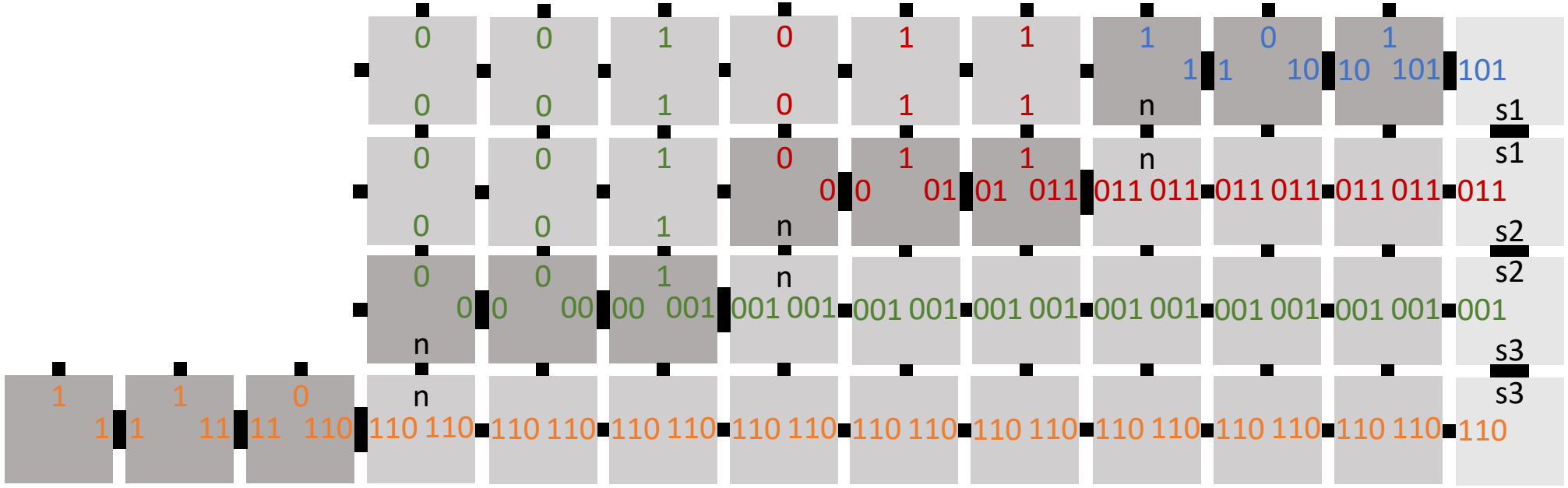

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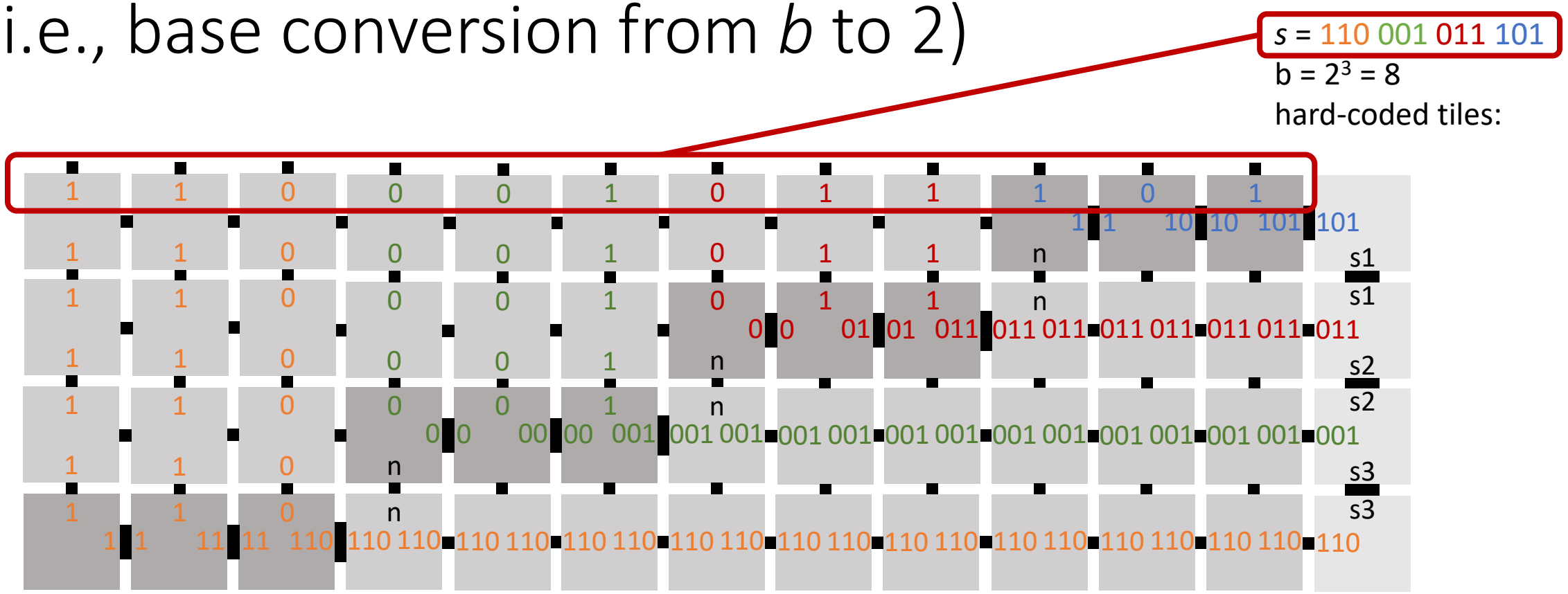

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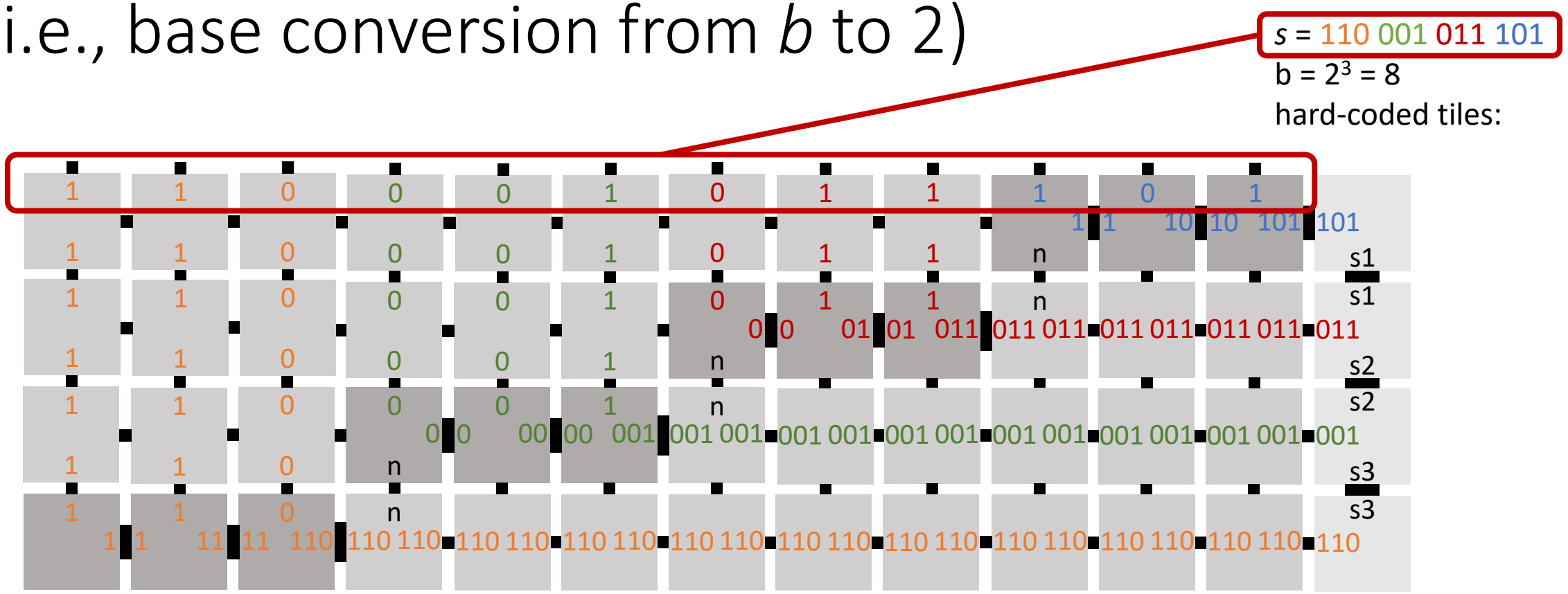

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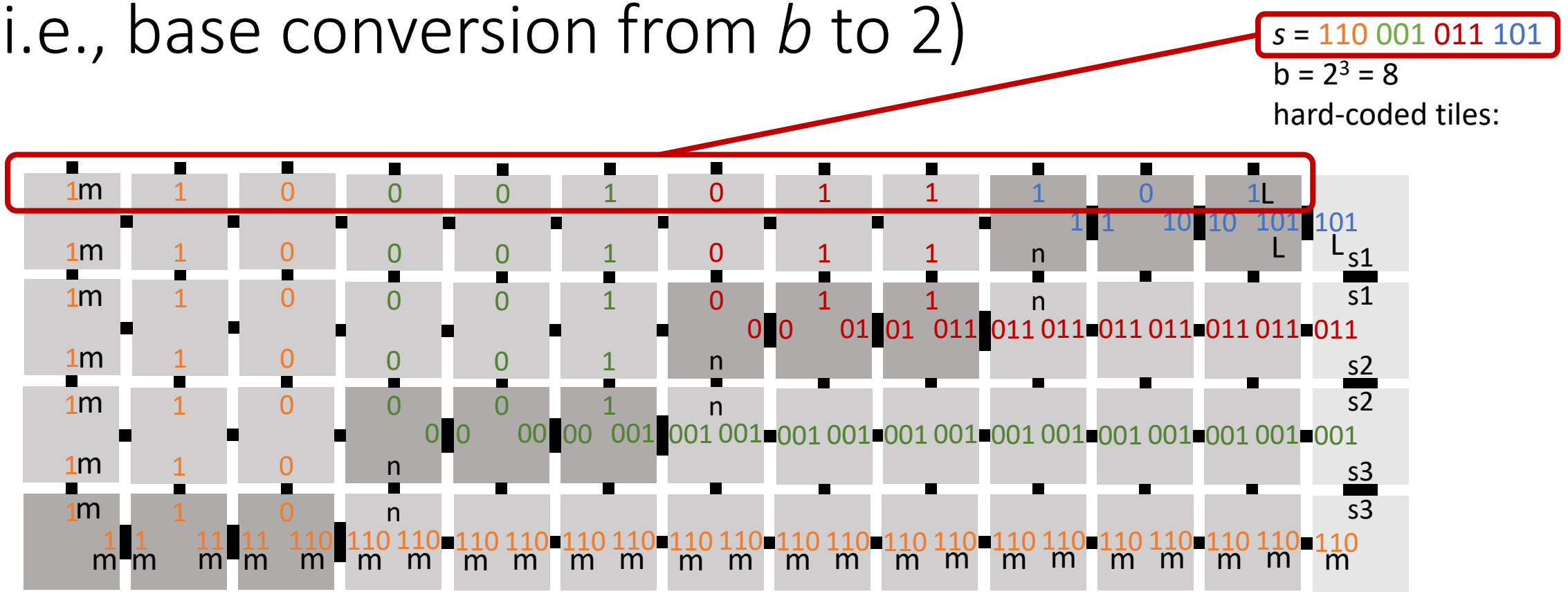

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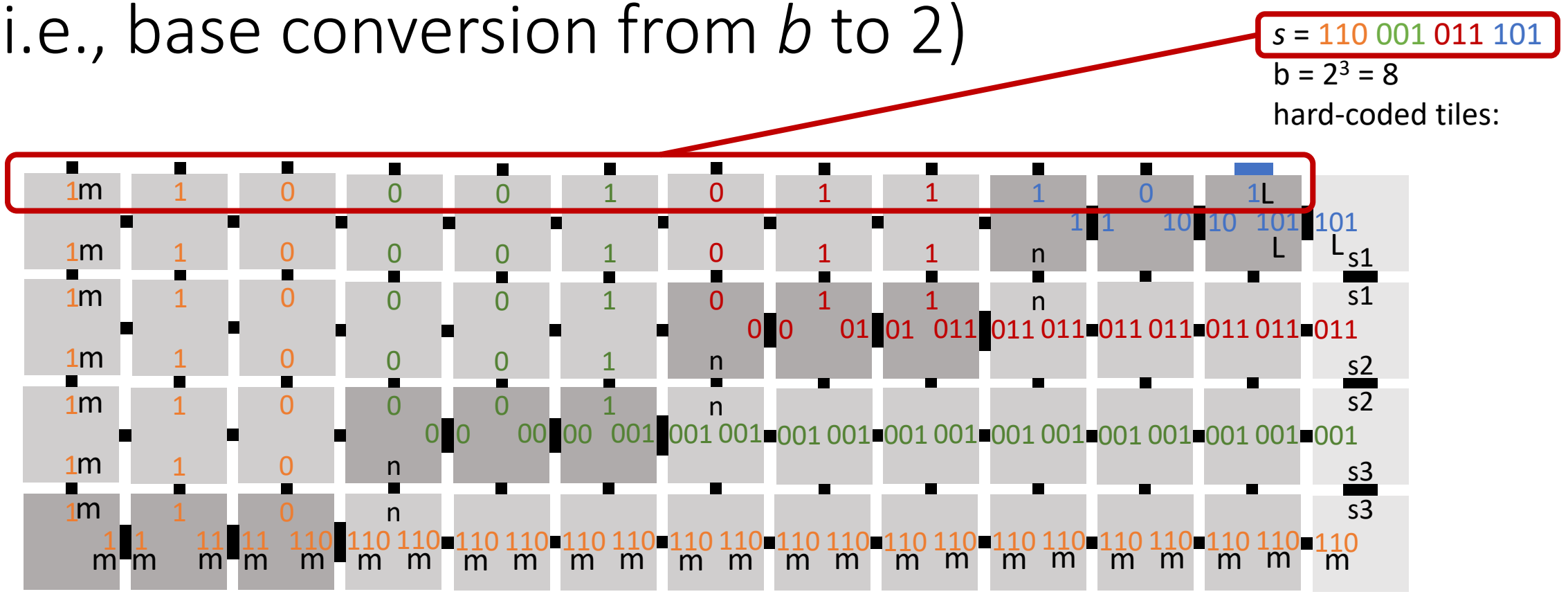

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Formal definition of aTAM

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Question: If $\alpha \sqsubseteq \beta$, can α grow into β?

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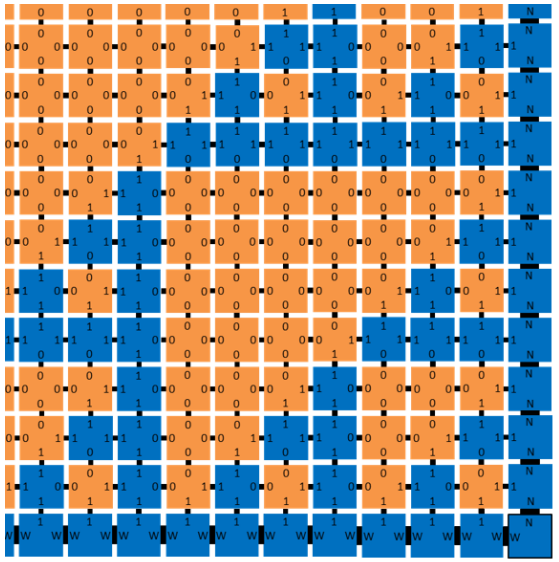

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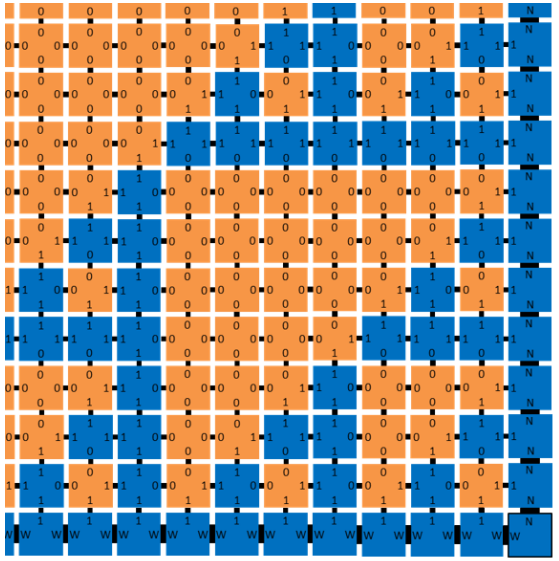

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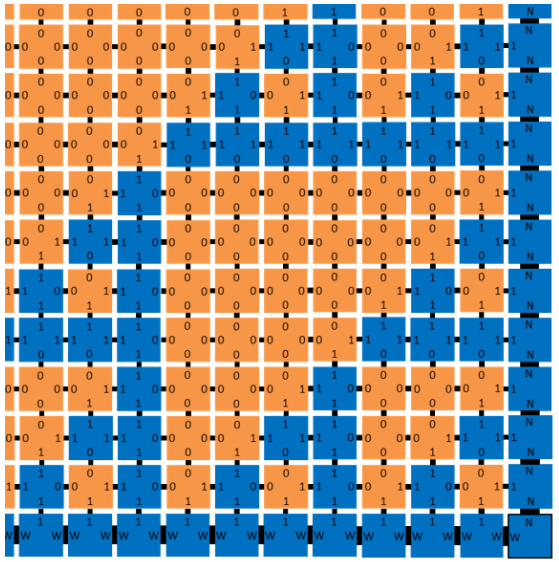

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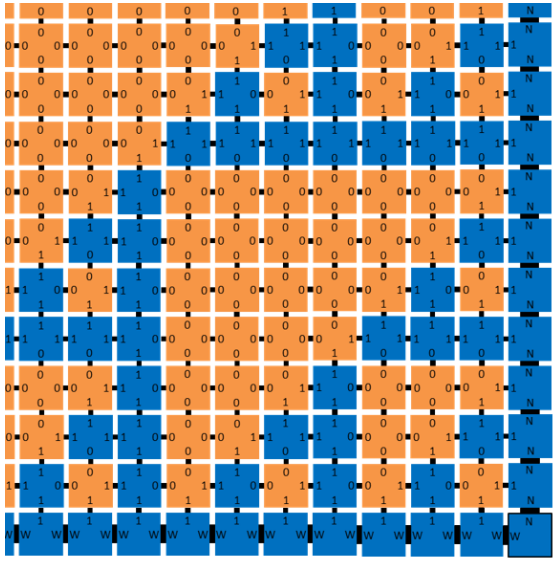

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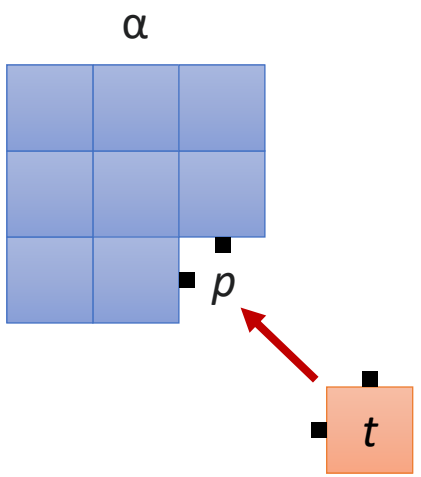

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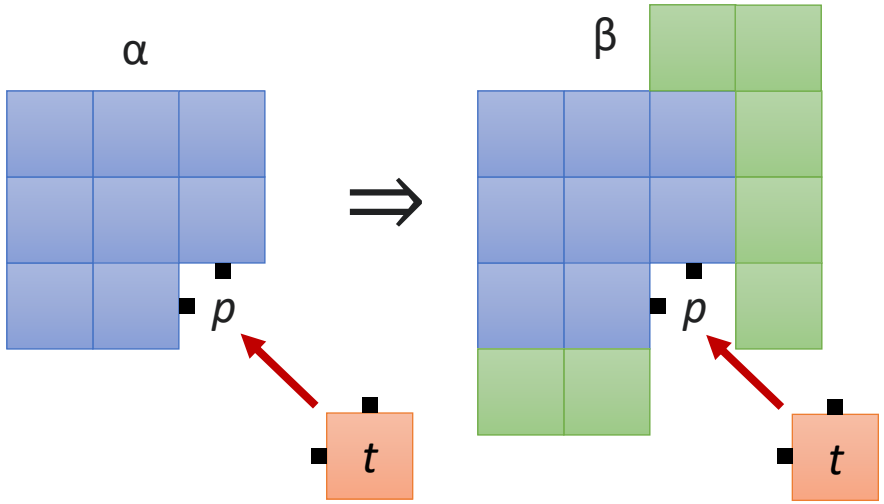

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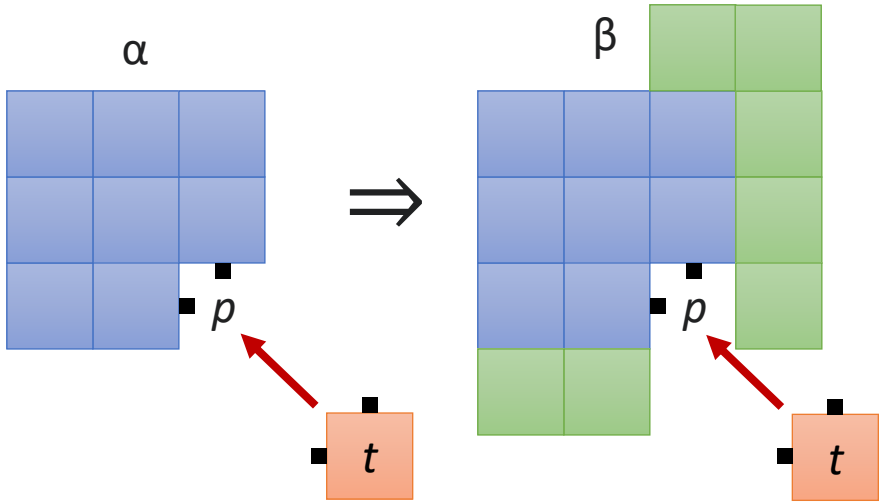

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1. Since β is stable and glue strengths are nonnegative, the only *potentially* unstable cut is $(\{p\}, S_\beta)$.

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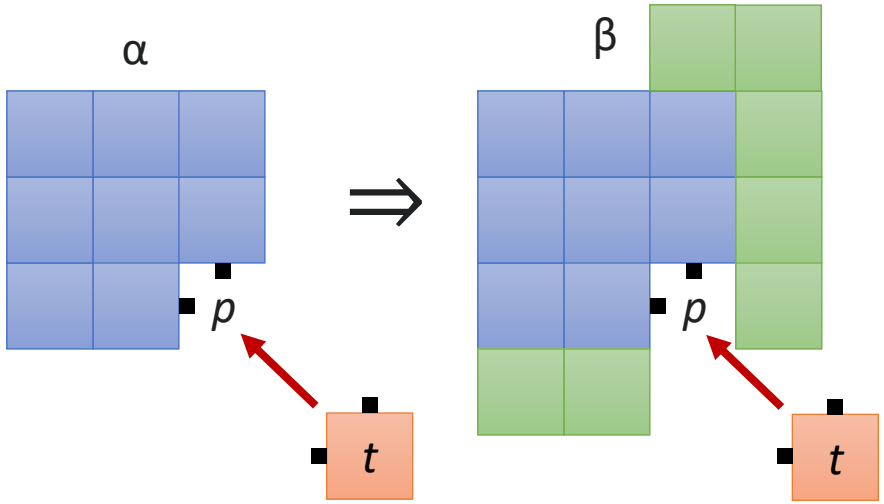

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Observation: Let $\alpha \sqsubseteq \beta$ be stable assemblies and $p \in \mathbb{Z}^2 \setminus S_\beta$ such that $\alpha + (p \mapsto t)$ is stable. Then $\beta + (p \mapsto t)$ is also stable.

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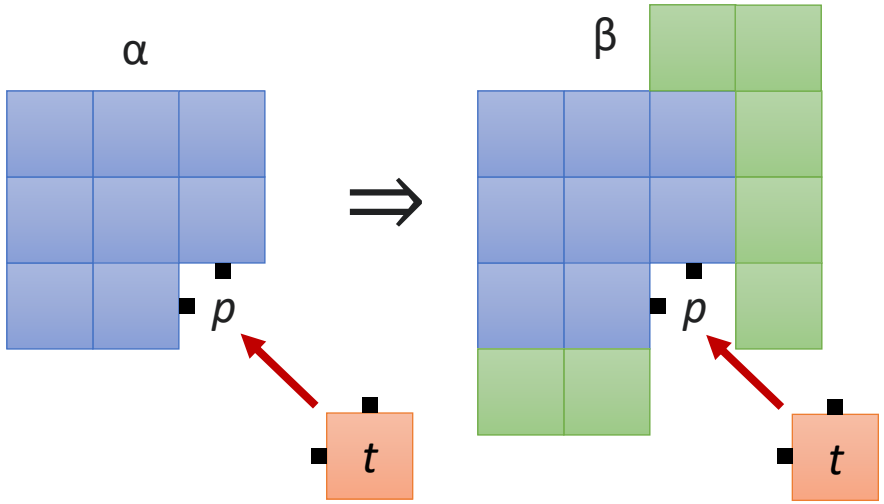

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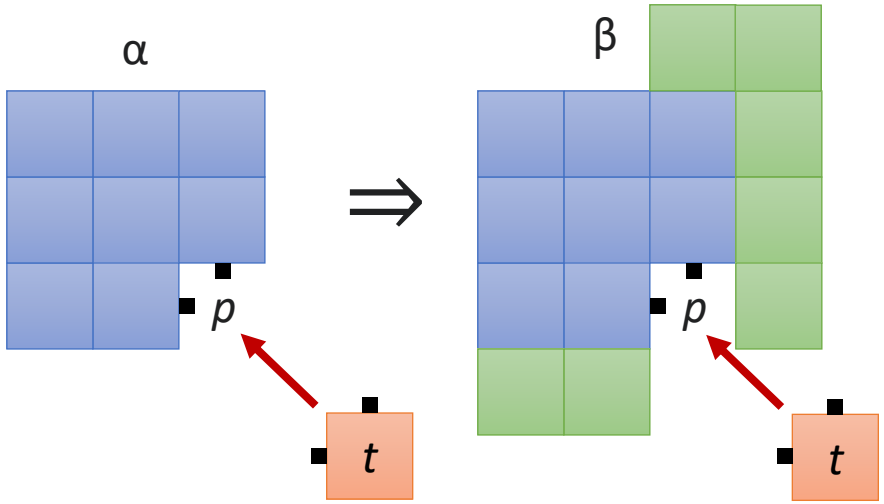

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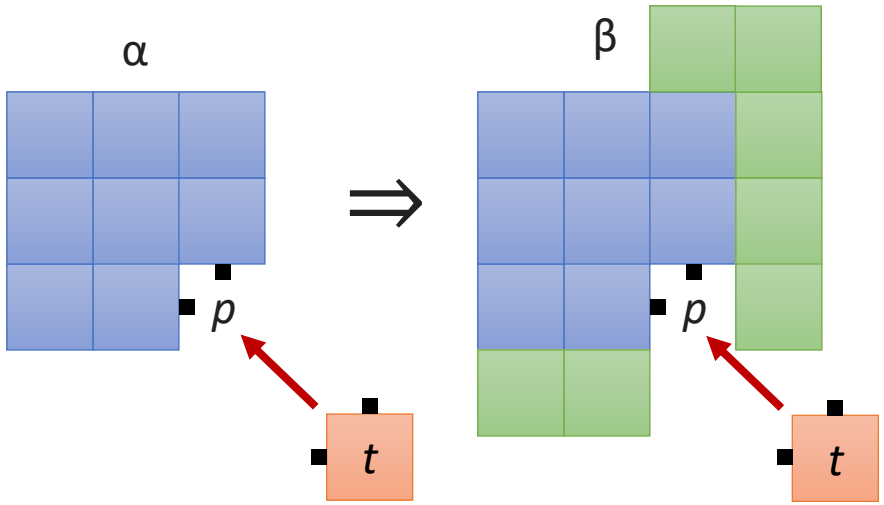

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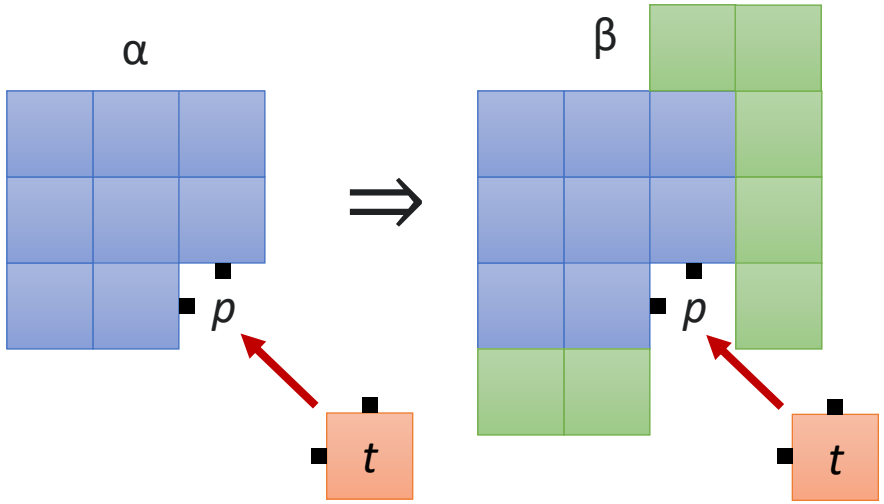

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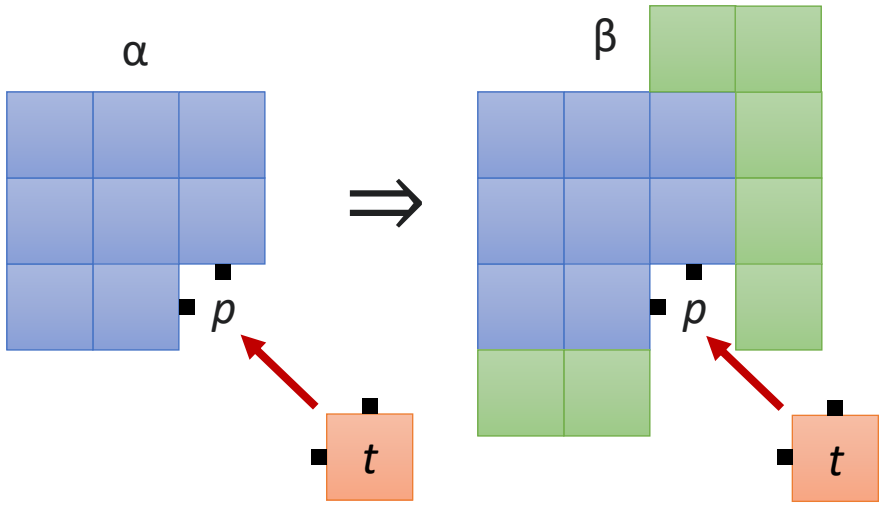

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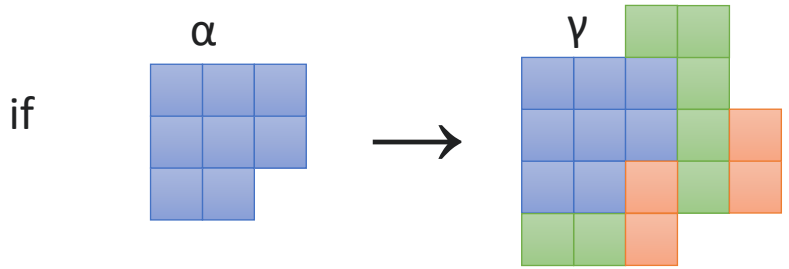

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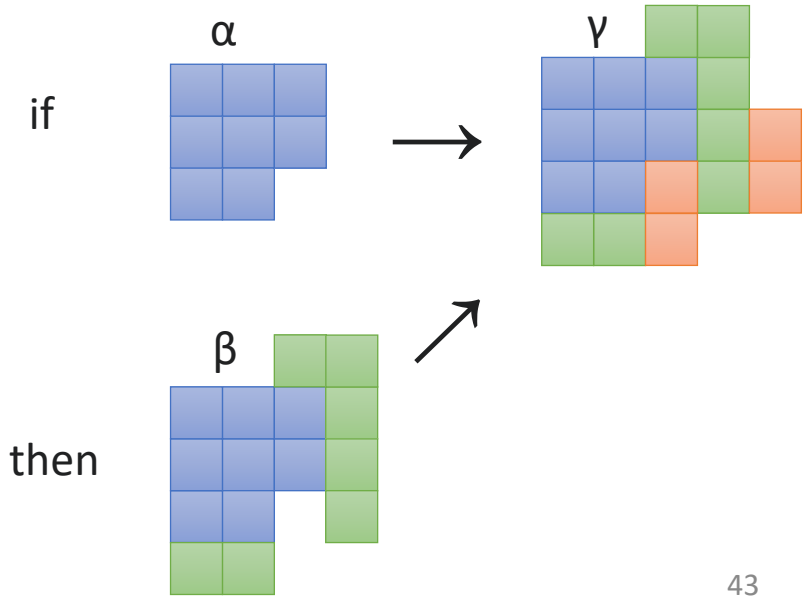

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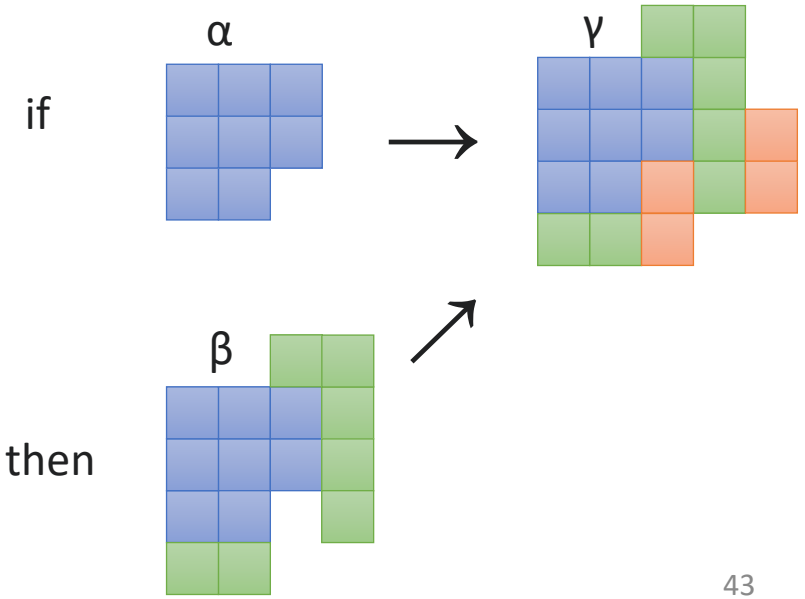

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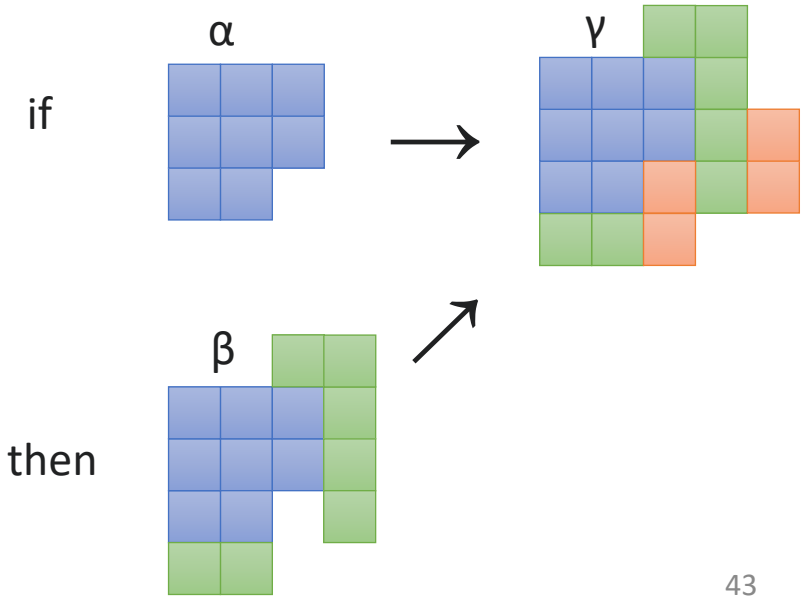

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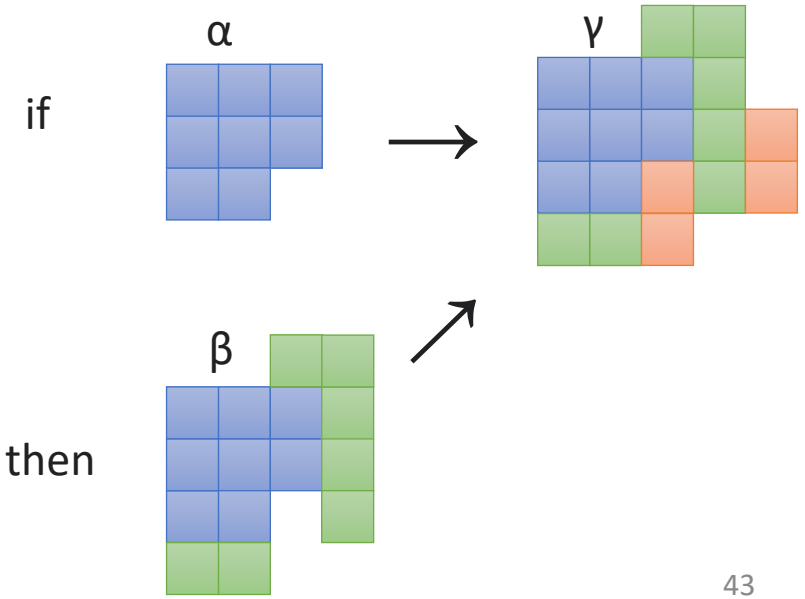

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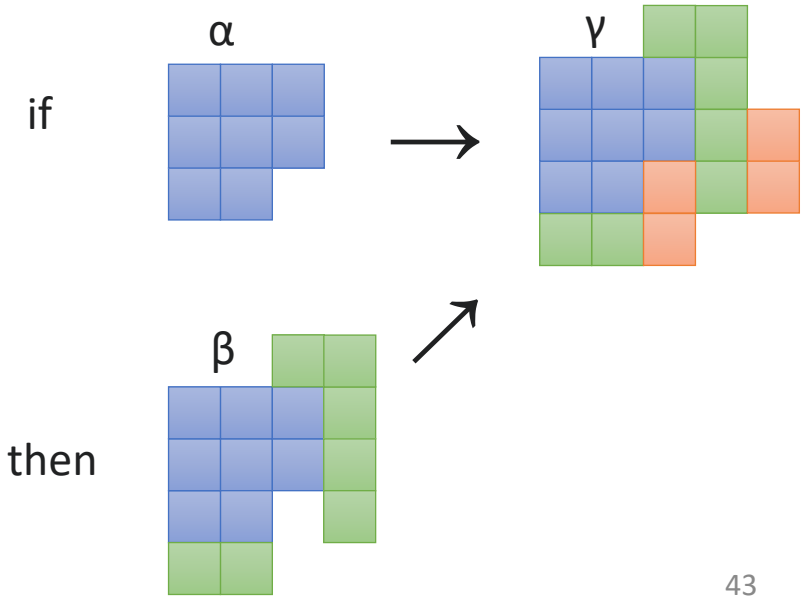

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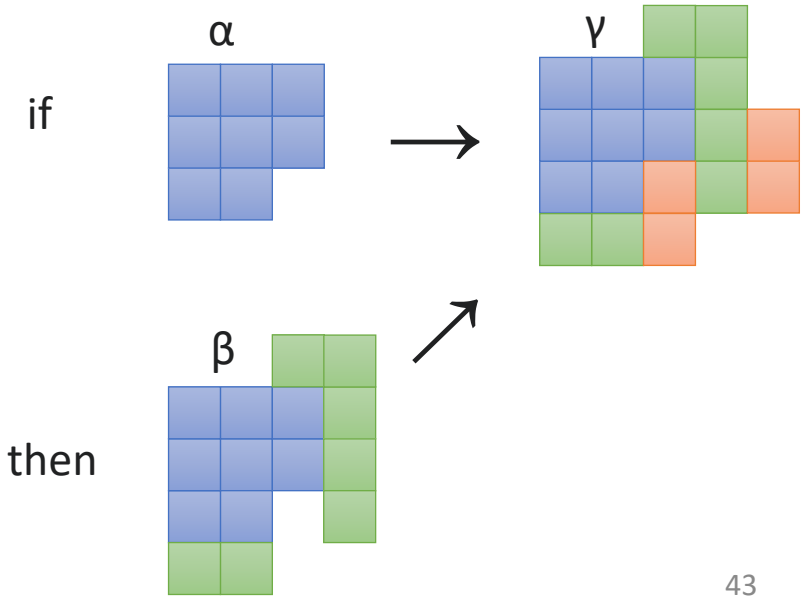

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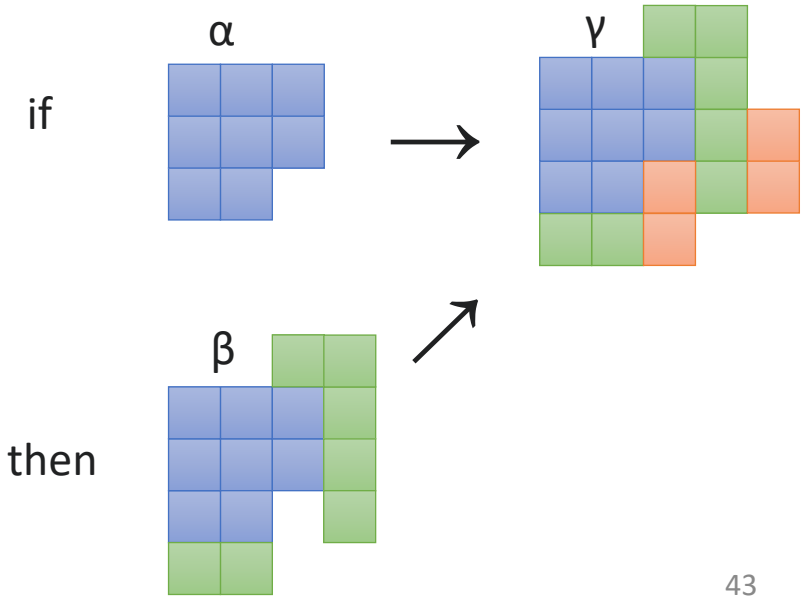

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6. Thus the assembly sequence is valid (each tile attachment is stable), showing $\beta \longrightarrow \gamma$. **QED**

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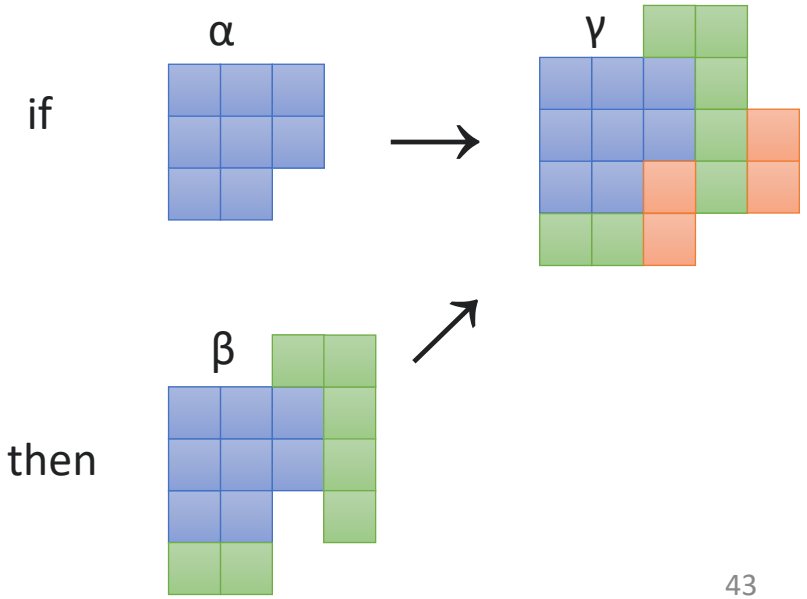

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- Recall two alternate characterizations of deterministic tile systems:

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- Rothemund's Lemma can be used to show that (b) implies (a)
  - will skip in lecture (optional problem on homework 1)

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3. Now assume assembly sequence is infinite. (*actually, rest of proof works in finite case*)
4. Since $p \in \partial\gamma$, there are positions adjacent to p with enough strength to bind a tile t. Let N be the set of these positions. Note N is finite since p has at most four neighbors.
5. Since $S_\gamma = \bigcup_i S_{\alpha i}$, there exists i such that $N \subseteq \partial\alpha_i$ (*after some finite number of tile attachments, all of the positions in N are on the frontier of the current assembly*)
6. Thus $p \in \partial\alpha_i$. (*the tile t can attach to α_i, reached after only i steps*)
7. By fairness, there exists j such that $p \in S_{\alpha j} \subseteq S_\gamma$ (*eventually p gets a tile*), which contradicts the claim that $p \notin S_\gamma$. **QED**

Fair assembly sequences

Definition: Let $\alpha_0, \alpha_1, \ldots$ be an assembly sequence. We say it is **fair** if, for all $i \in \mathbb{N}$ and all $p \in \partial\alpha_i$, there exists $j > i$ such that $p \in S_{\alpha j}$.

Intuition: Every frontier location eventually gets a tile; none are "starved"

Lemma: Let $\alpha_0, \alpha_1, \ldots$ be a fair assembly sequence. Then its result γ is terminal.

Corollary: For every assembly α, there is a terminal assembly γ such that $\alpha \longrightarrow \gamma$.

Proof:

1. Suppose for the sake of contradiction that γ is not terminal, i.e., it has frontier location $p \in \partial\gamma$; note in particular $p \notin S_\gamma$.
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Proof: Pick any fair assembly sequence $\alpha=\alpha_0, \alpha_1, \ldots$; its result γ is terminal and $\alpha \longrightarrow \gamma$. **QED**

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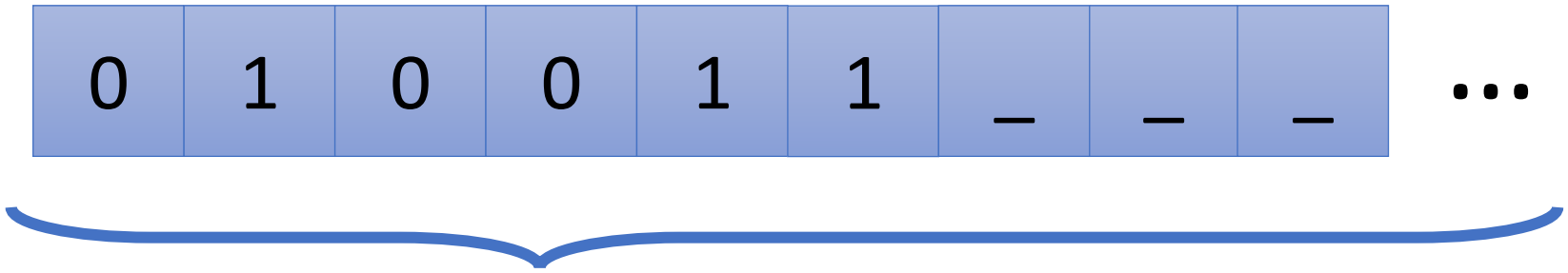
Concrete example of simulation algorithm creating a fair assembly sequence?

How computationally powerful are self-assembling tiles?

Turing machines

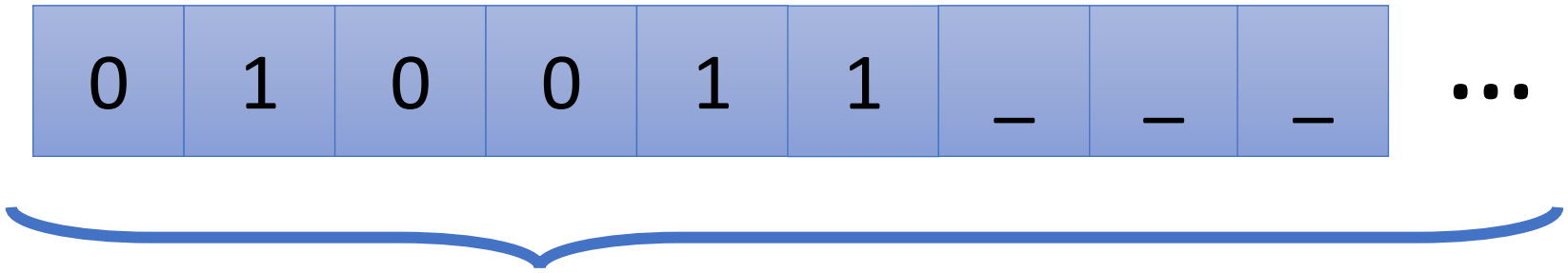
Turing machines



tape ≈ memory

Turing machines

state ≈ line of code



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Turing machines

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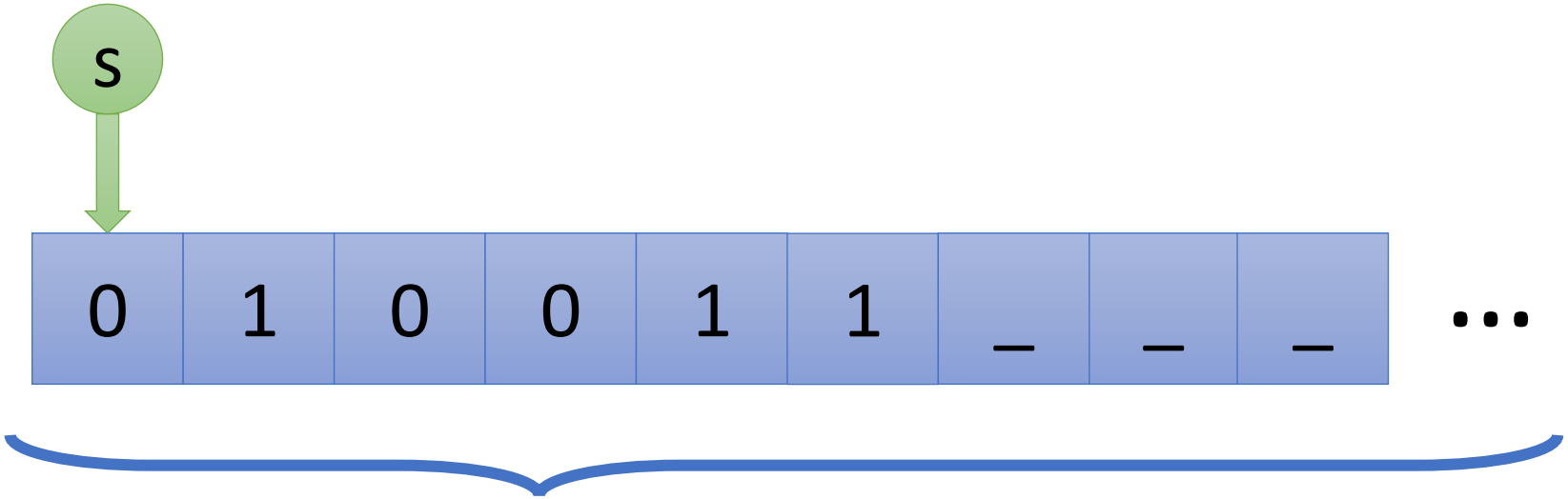
initial state = s



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Turing machines

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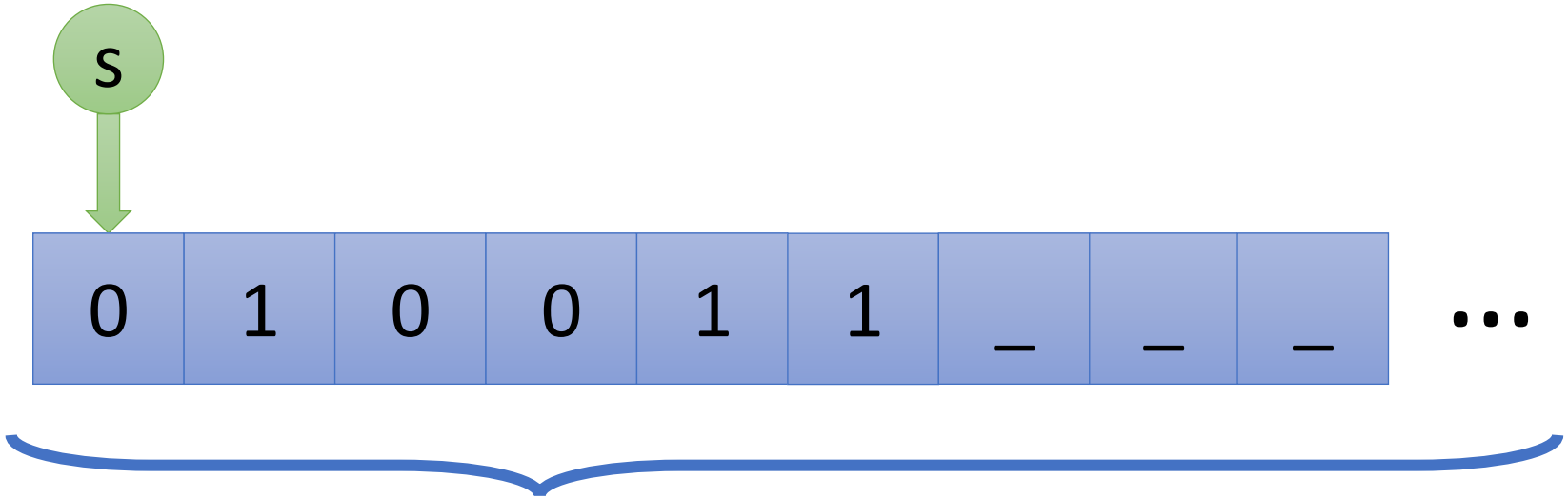
initial state = s

s,0: q,0,→



transitions
(instructions)

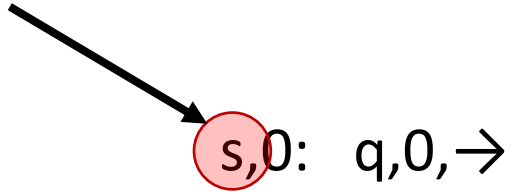
tape ≈ memory

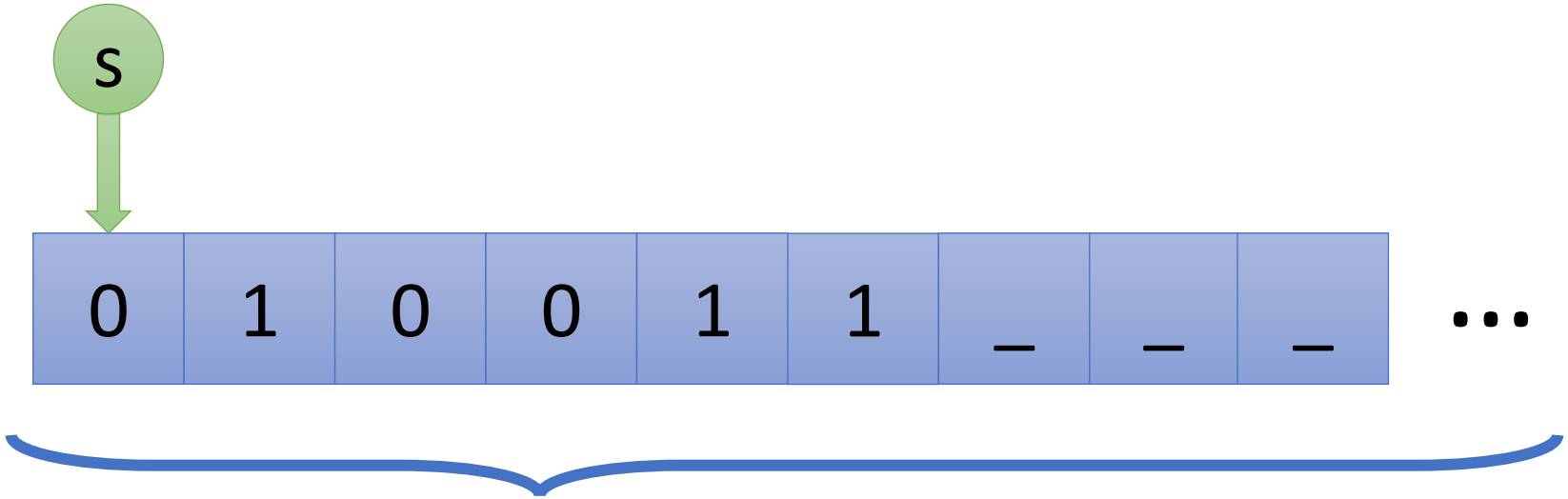
Turing machines

state ≈ line of code

initial state = s

current state





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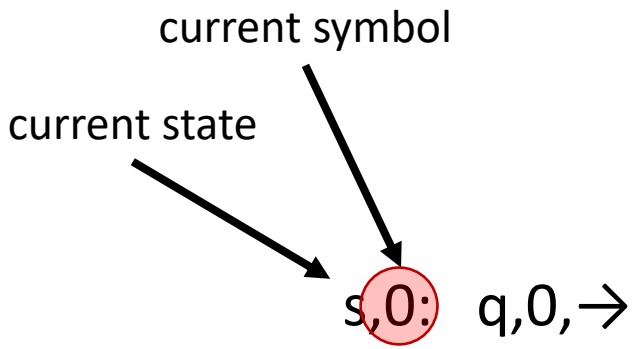

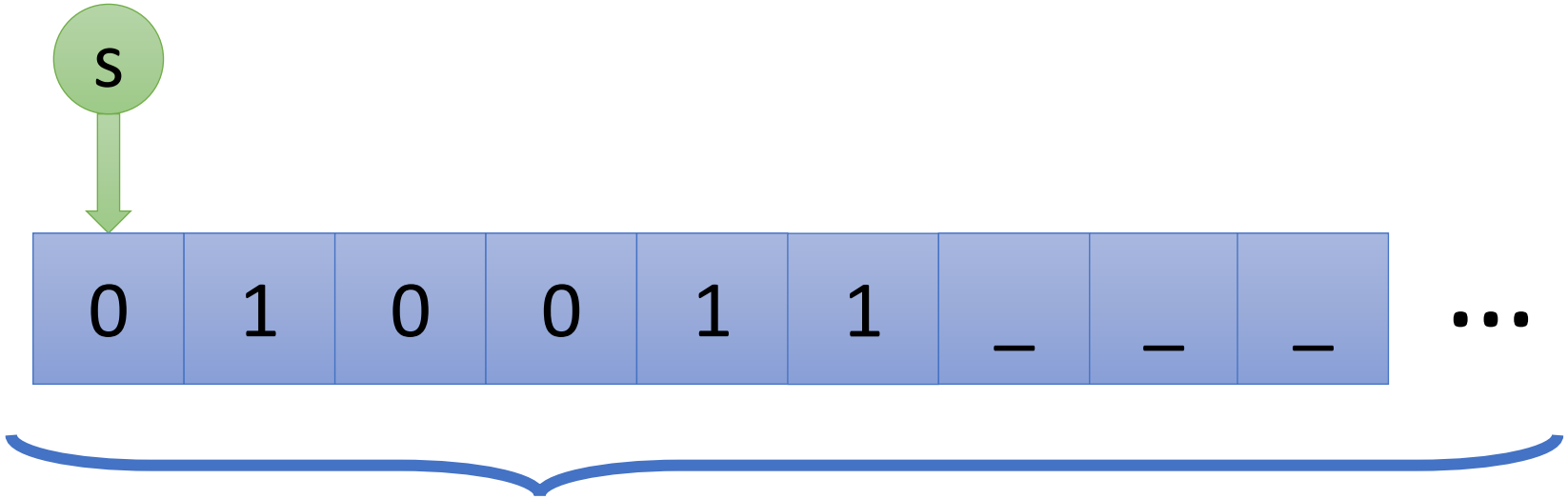

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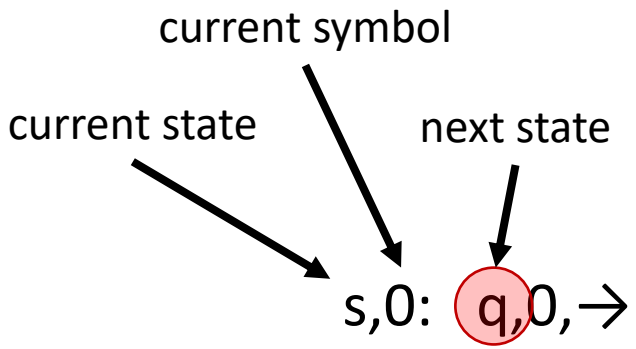

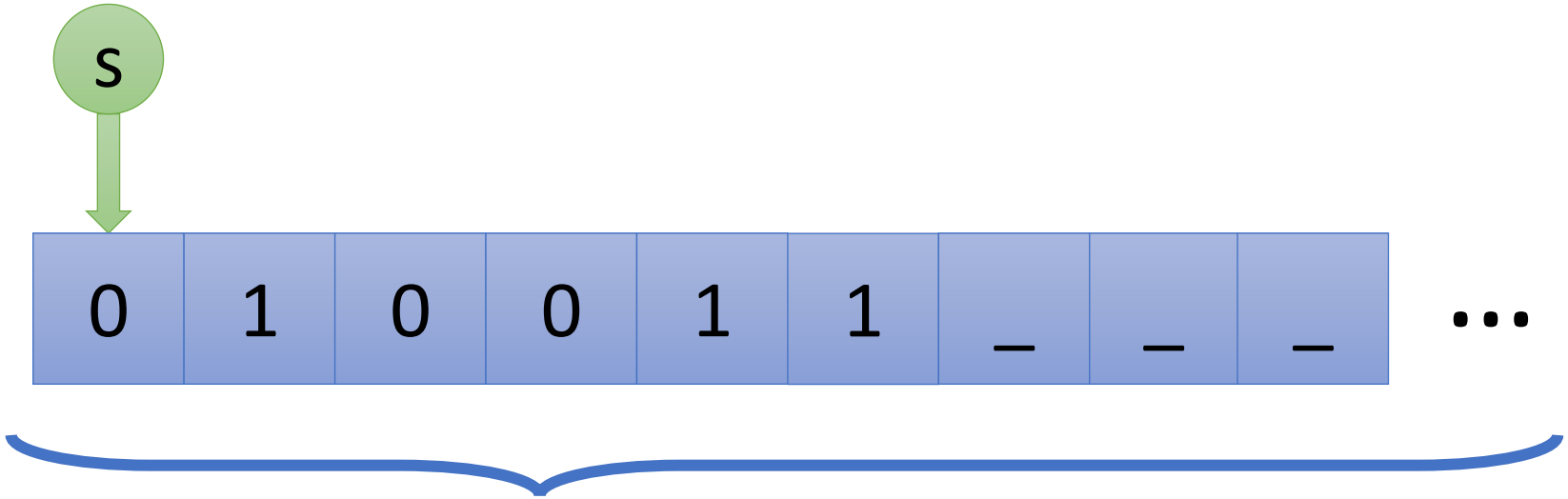

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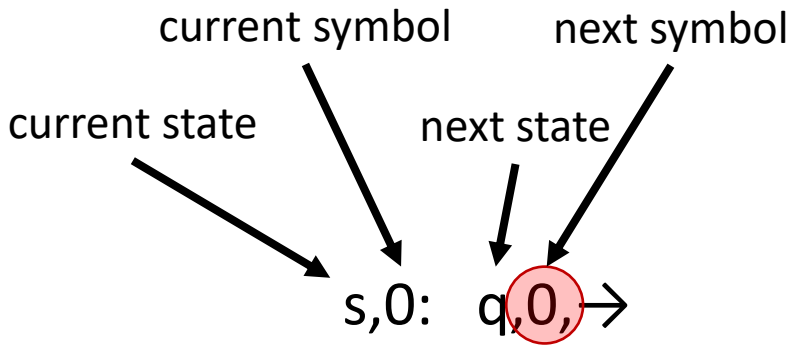

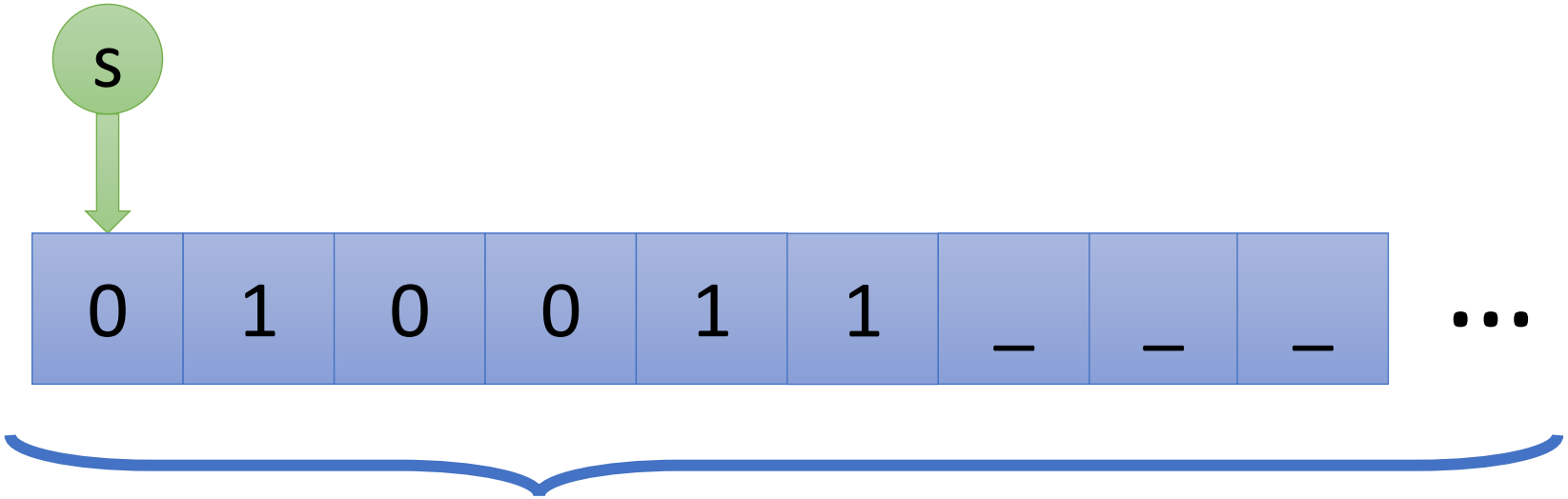

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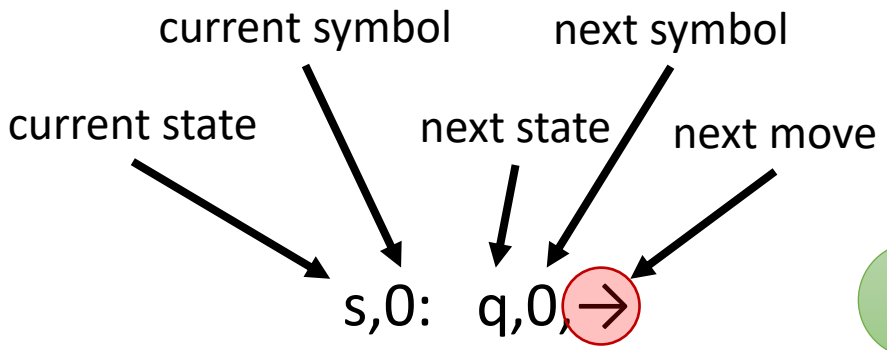

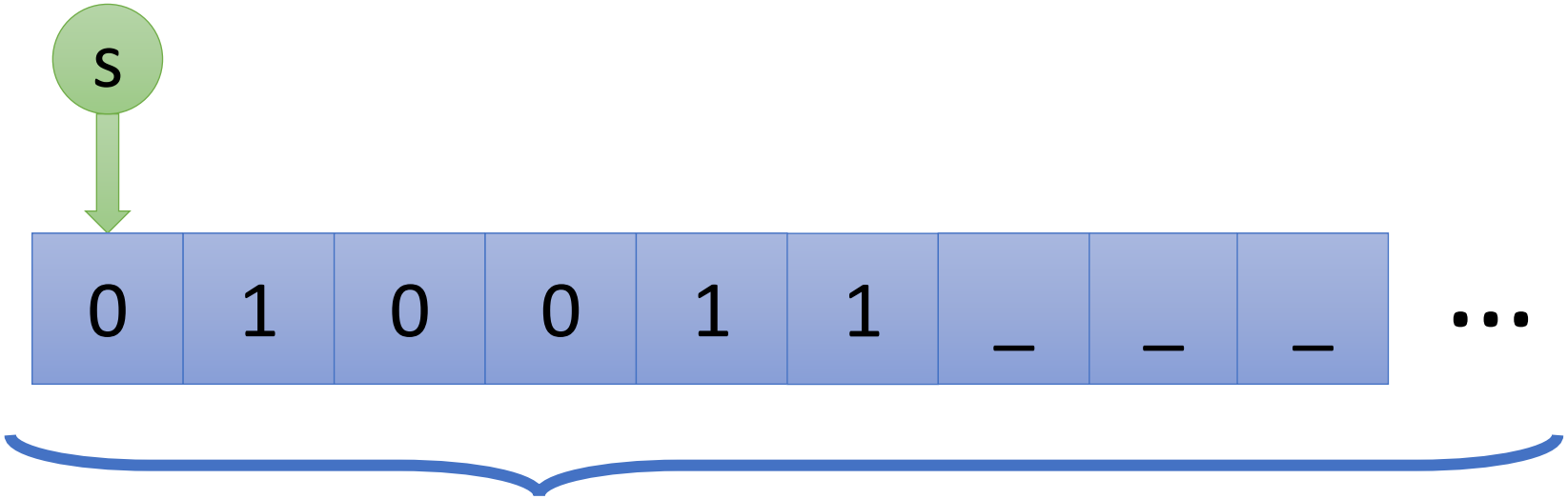

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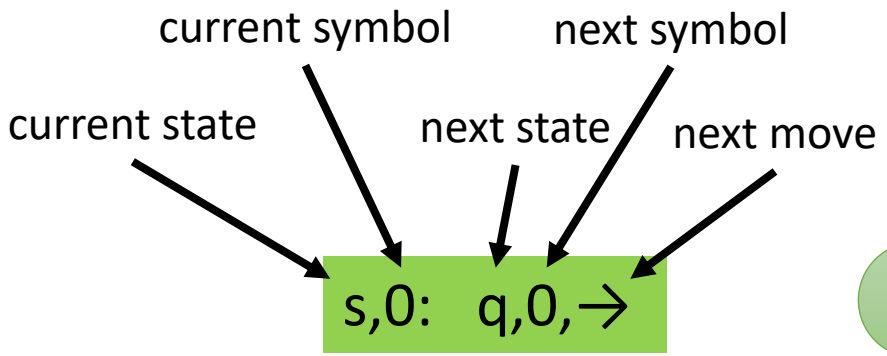


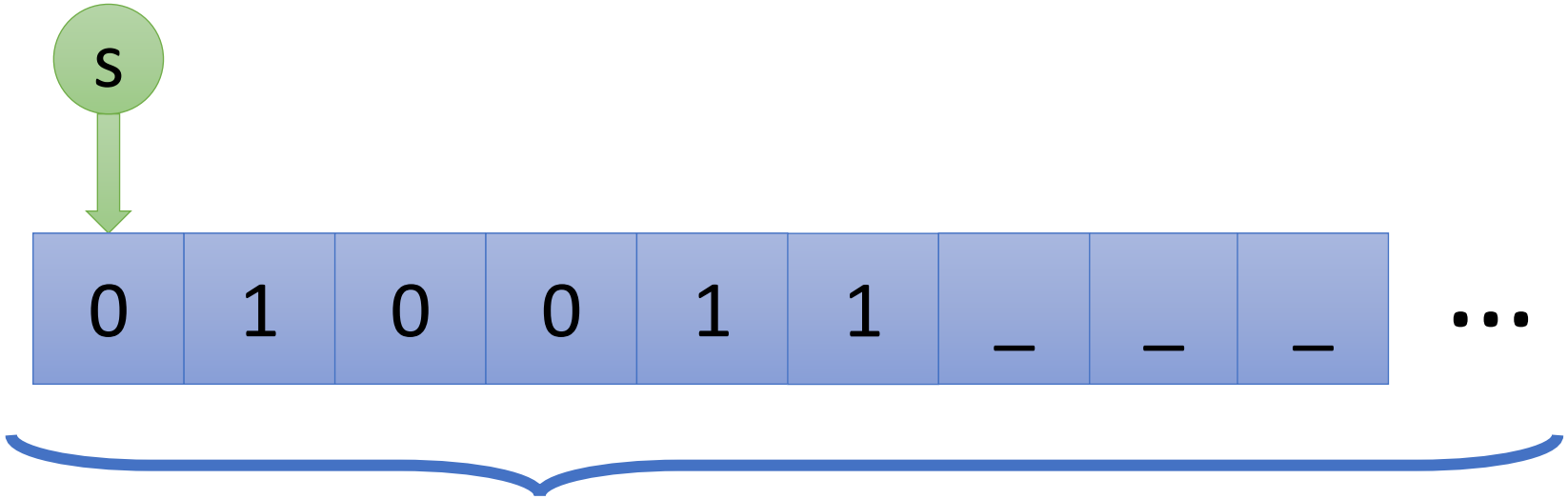
q,0: t,1,←

q,1: s,0,→

t,0: u,1,→

u,1: HALT

transitions
(instructions)



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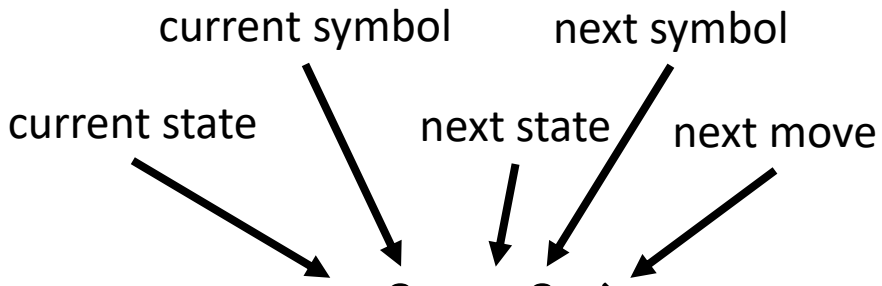

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transitions

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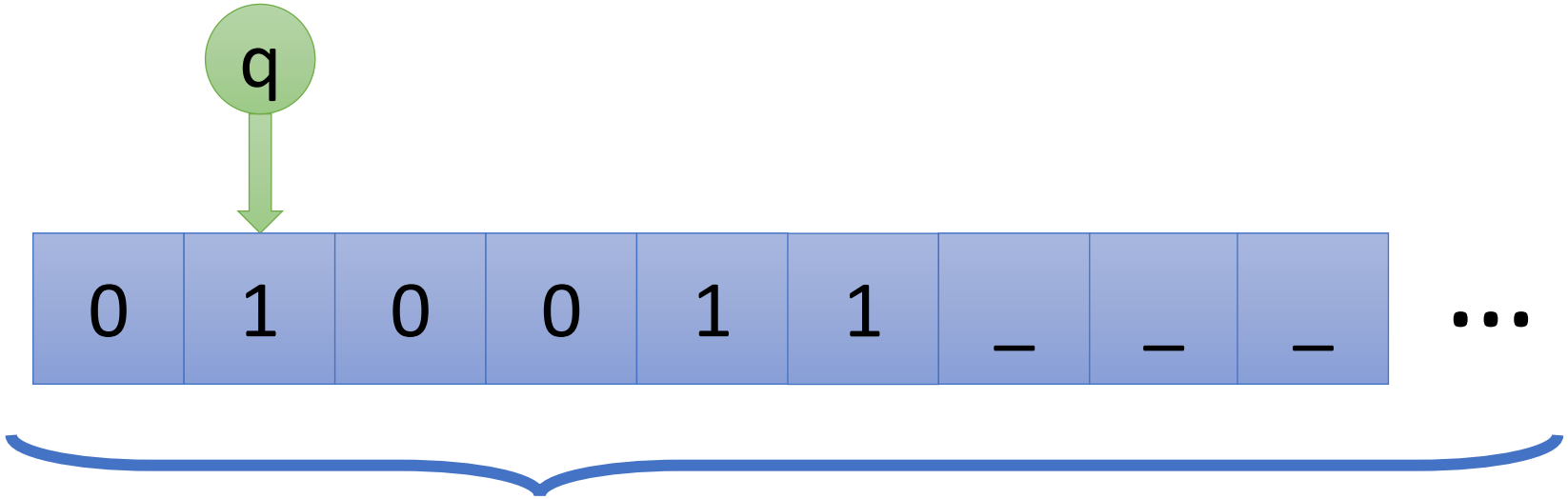

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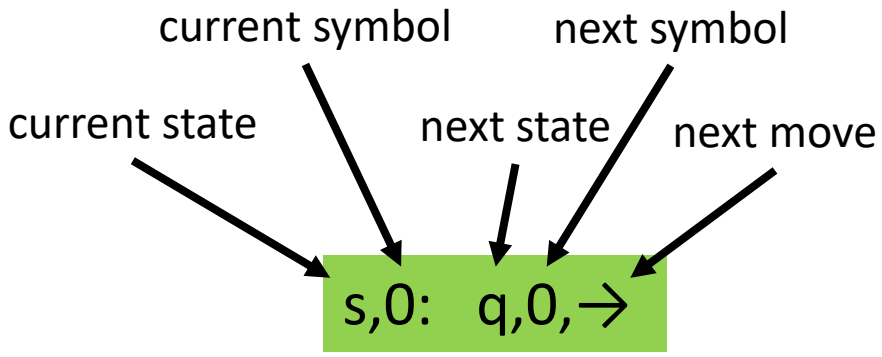

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transitions
(instructions)

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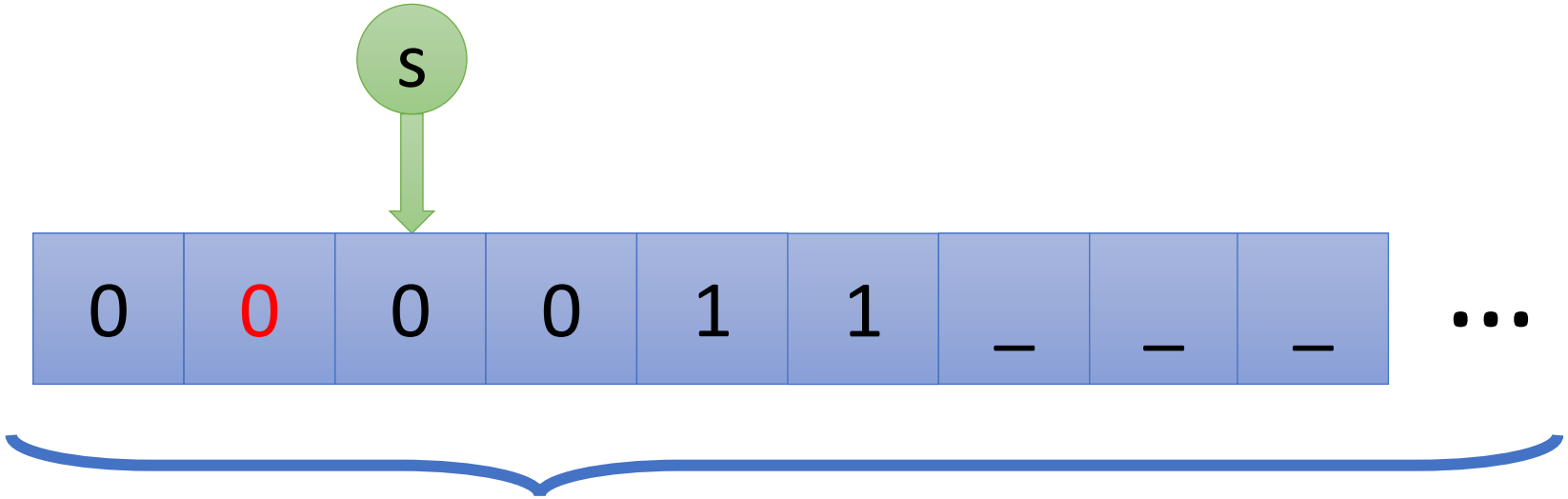

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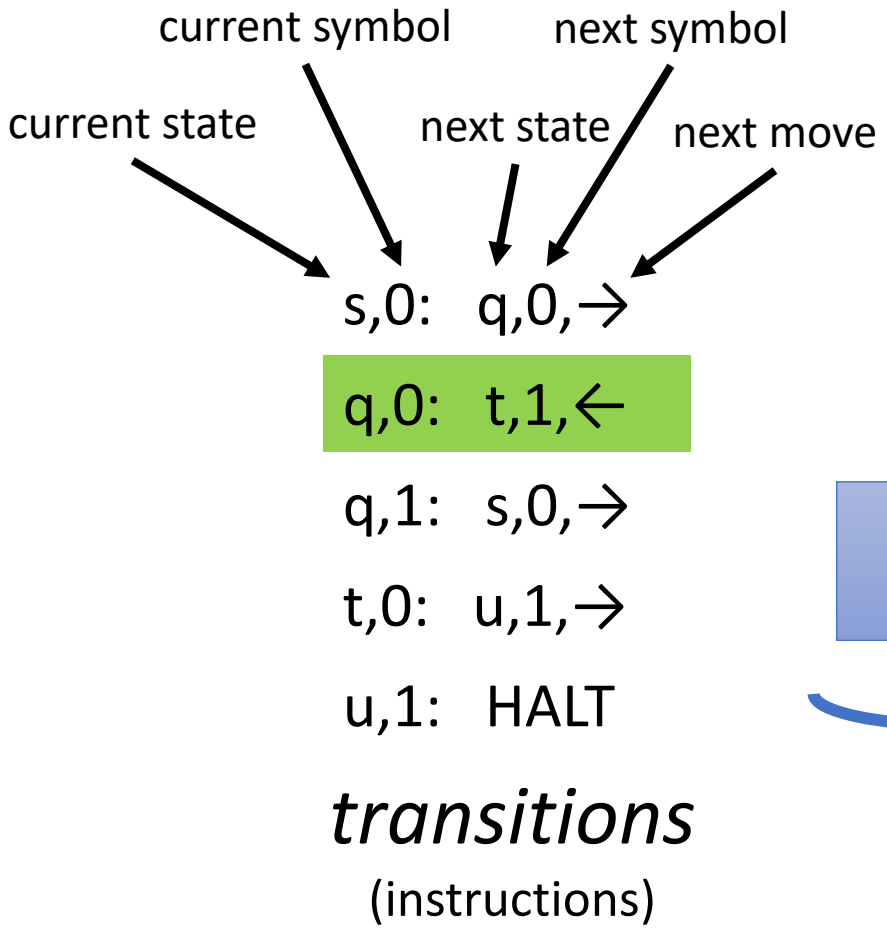

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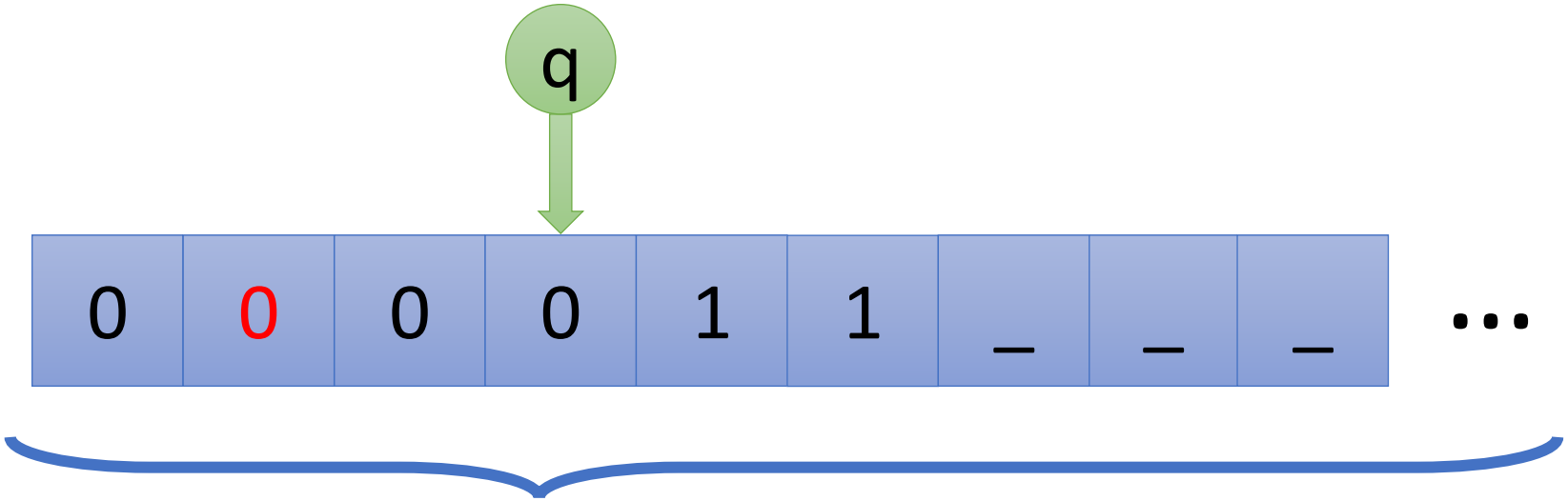

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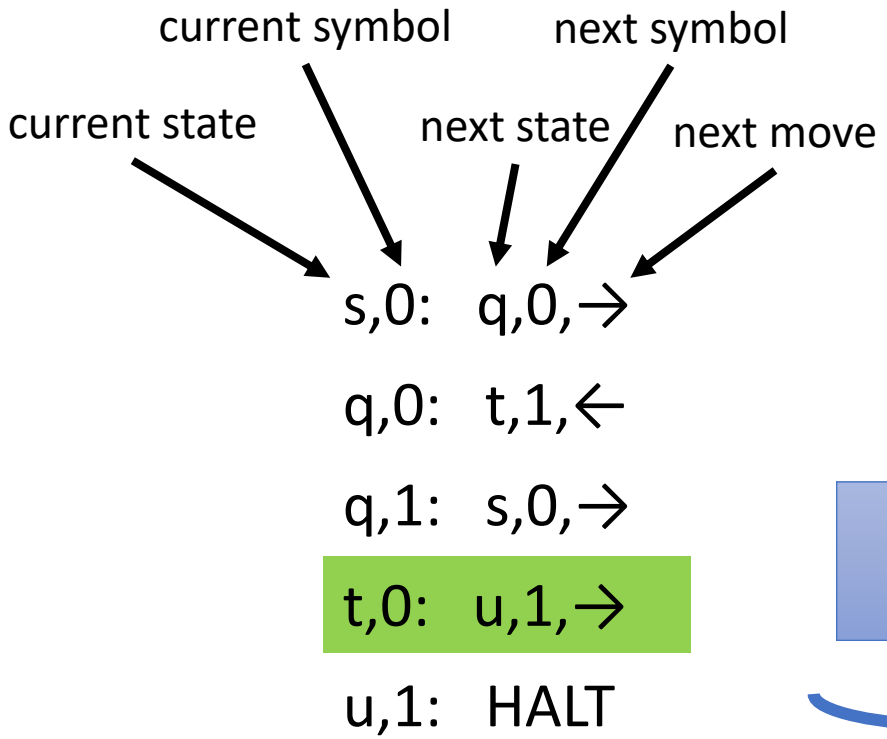
Turing machines



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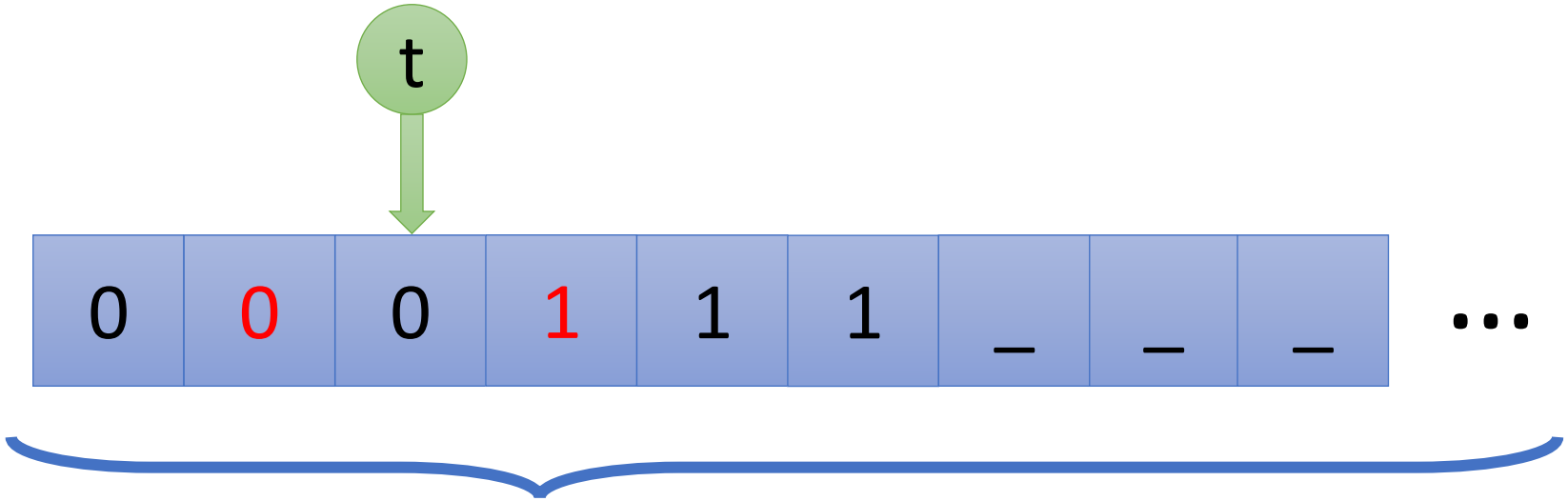

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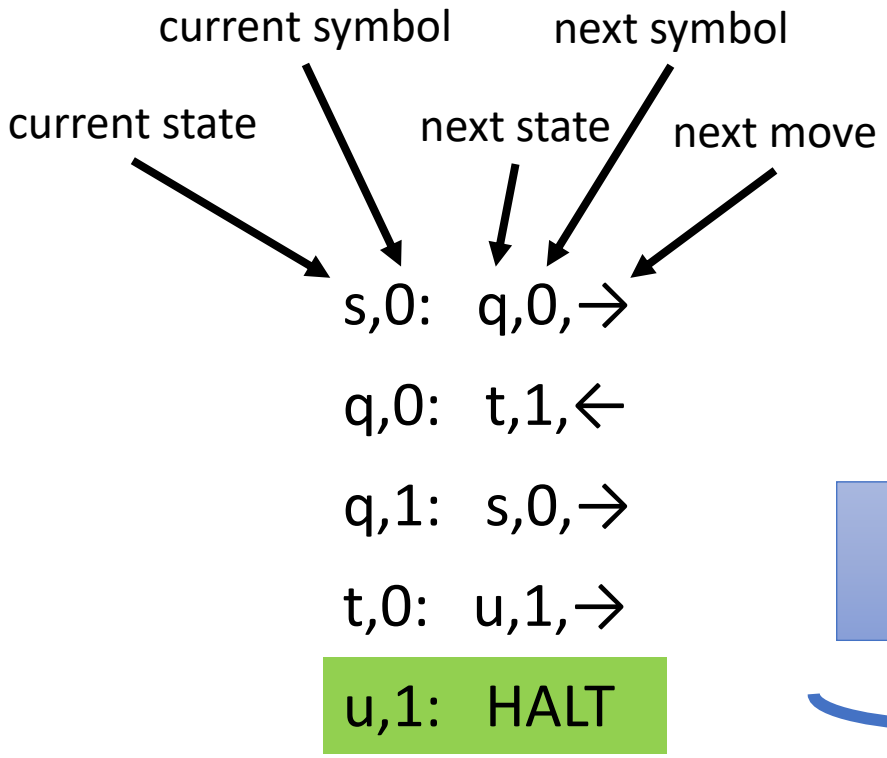

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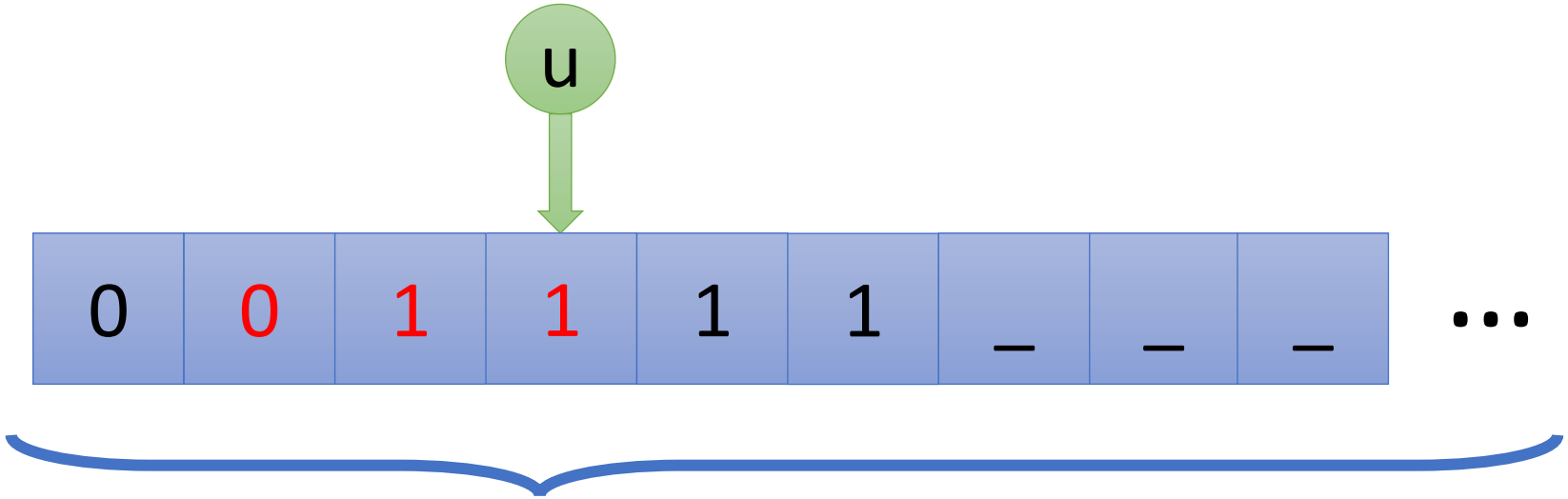

tape ≈ memory

Tile assembly is Turing-universal

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q,0: t,1,←

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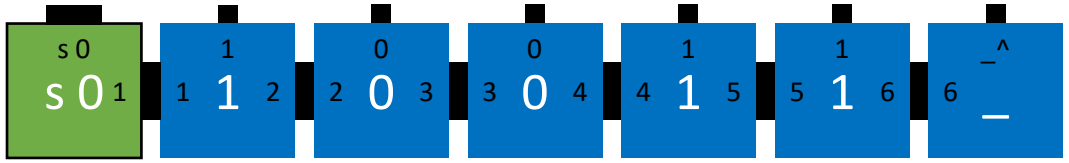

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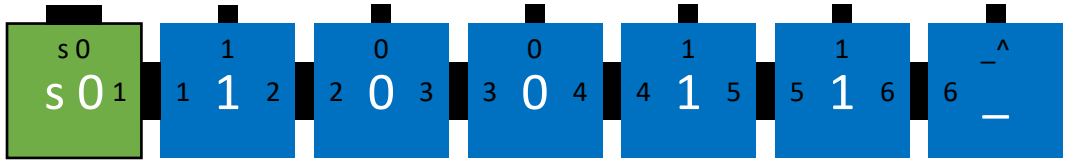

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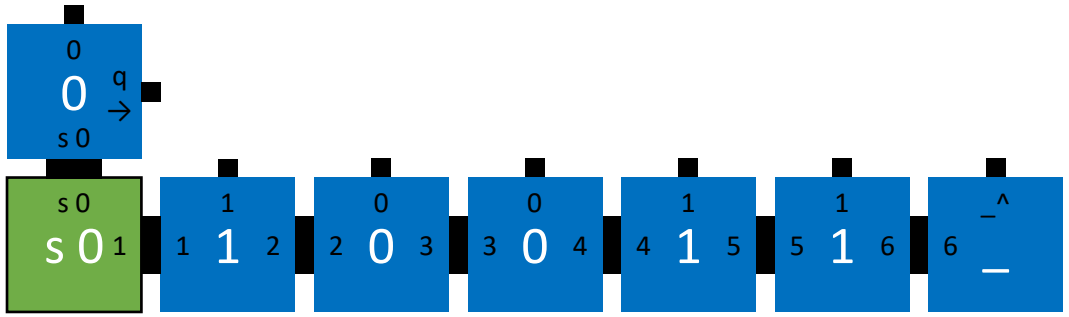

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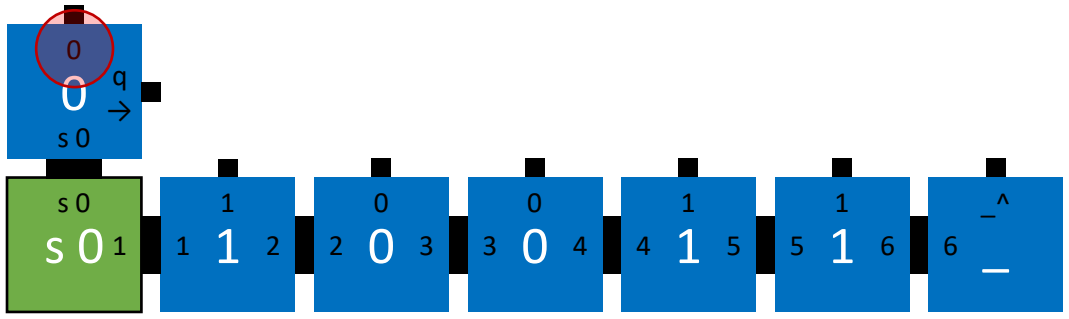

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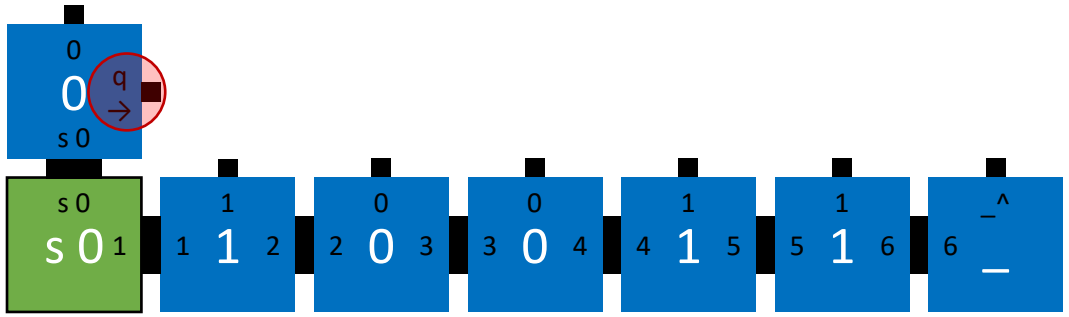

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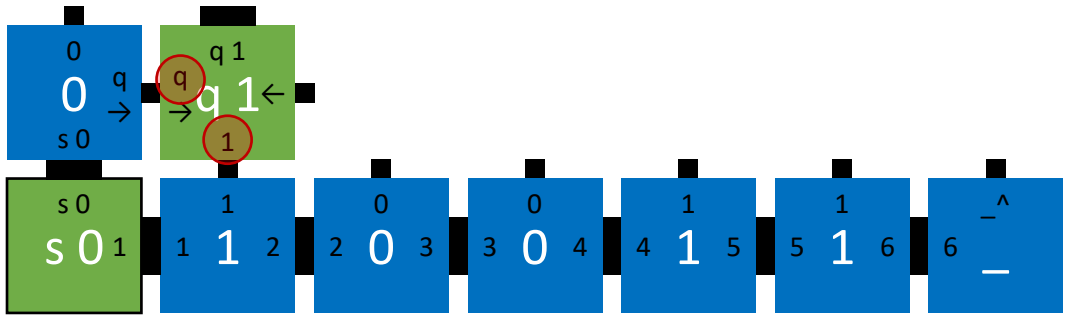

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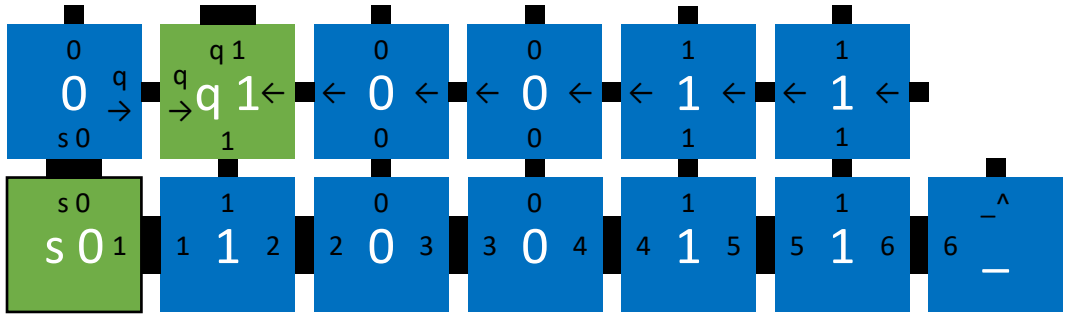

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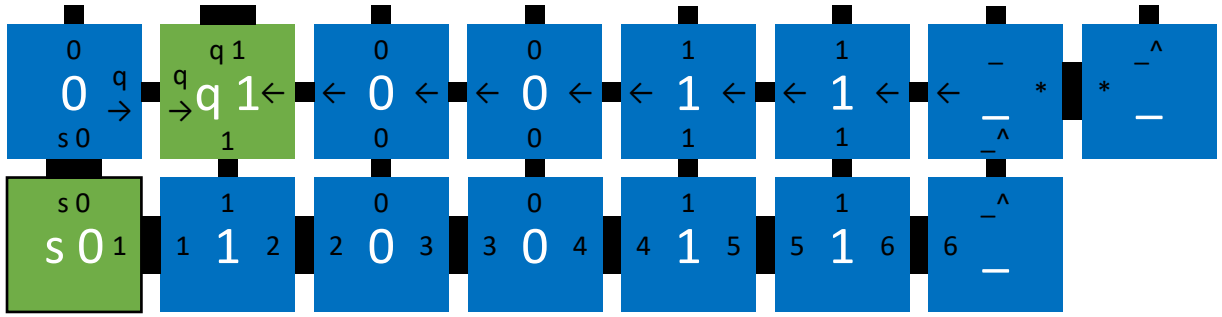

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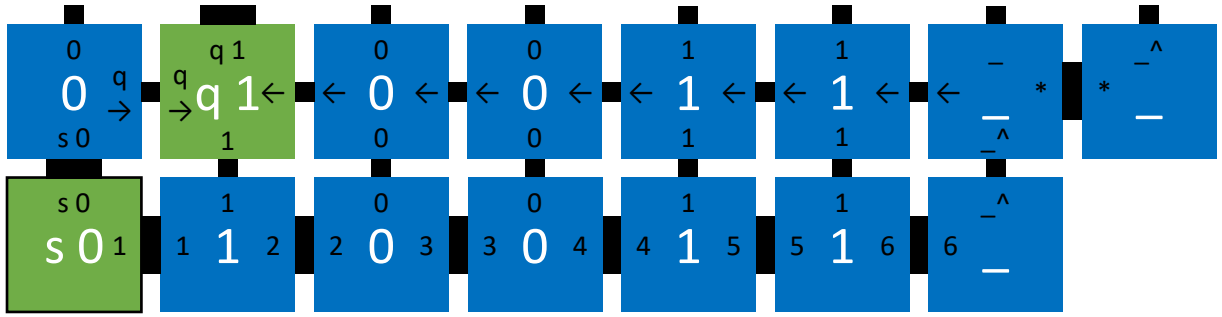
q,1: s,0,→

t,0: u,1,→

u,1: HALT

Tile assembly is Turing-universal



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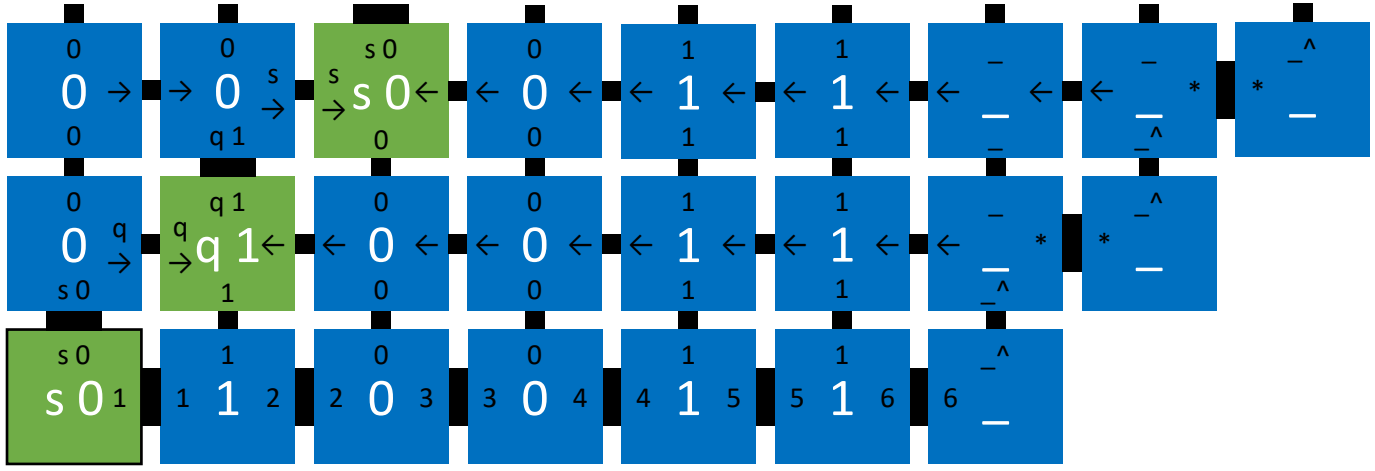

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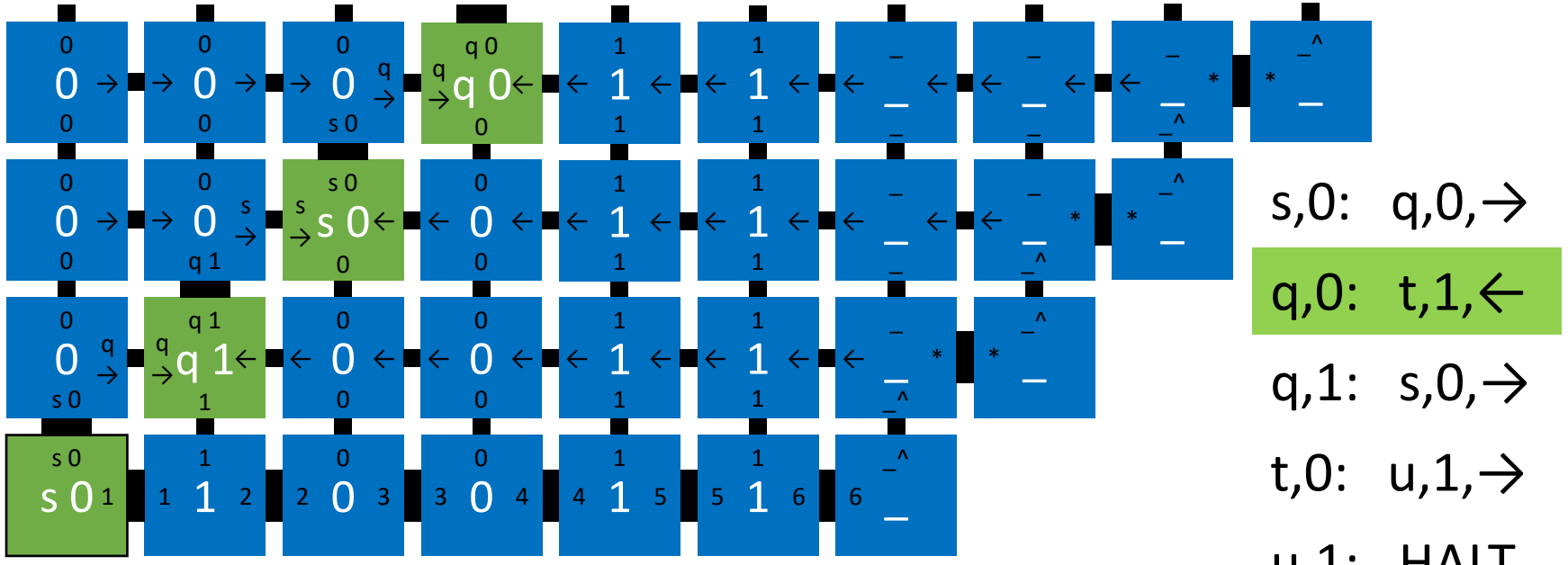

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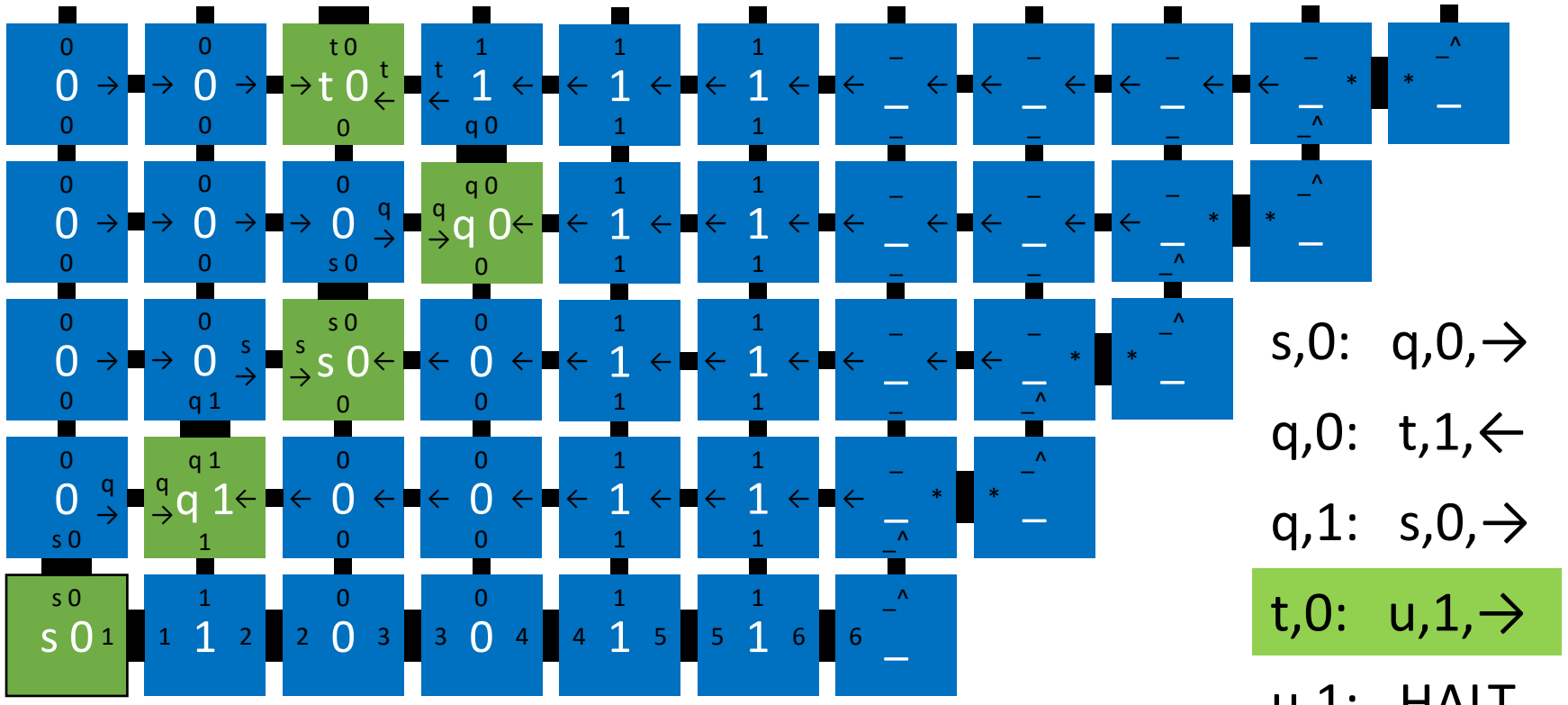

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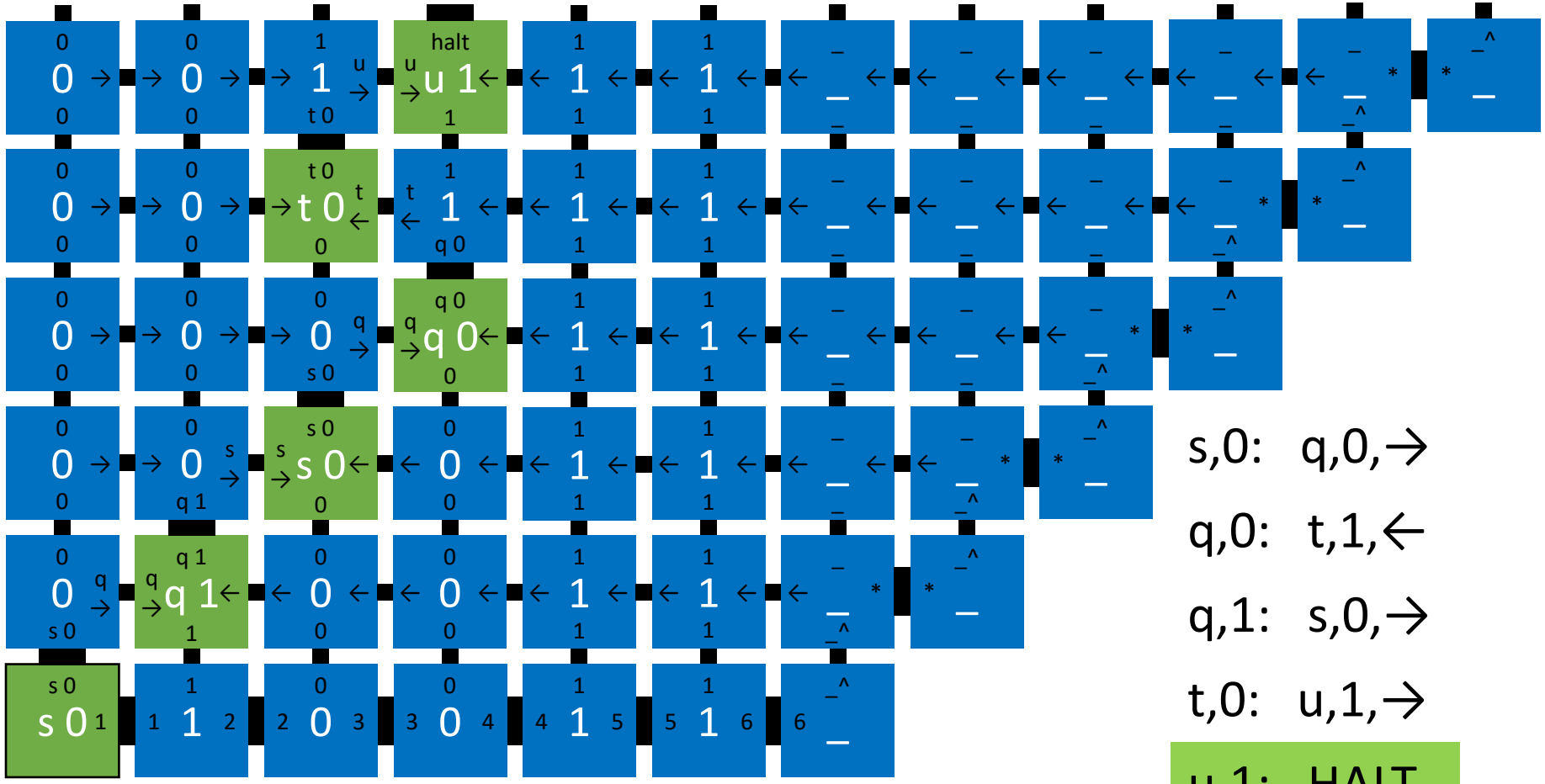

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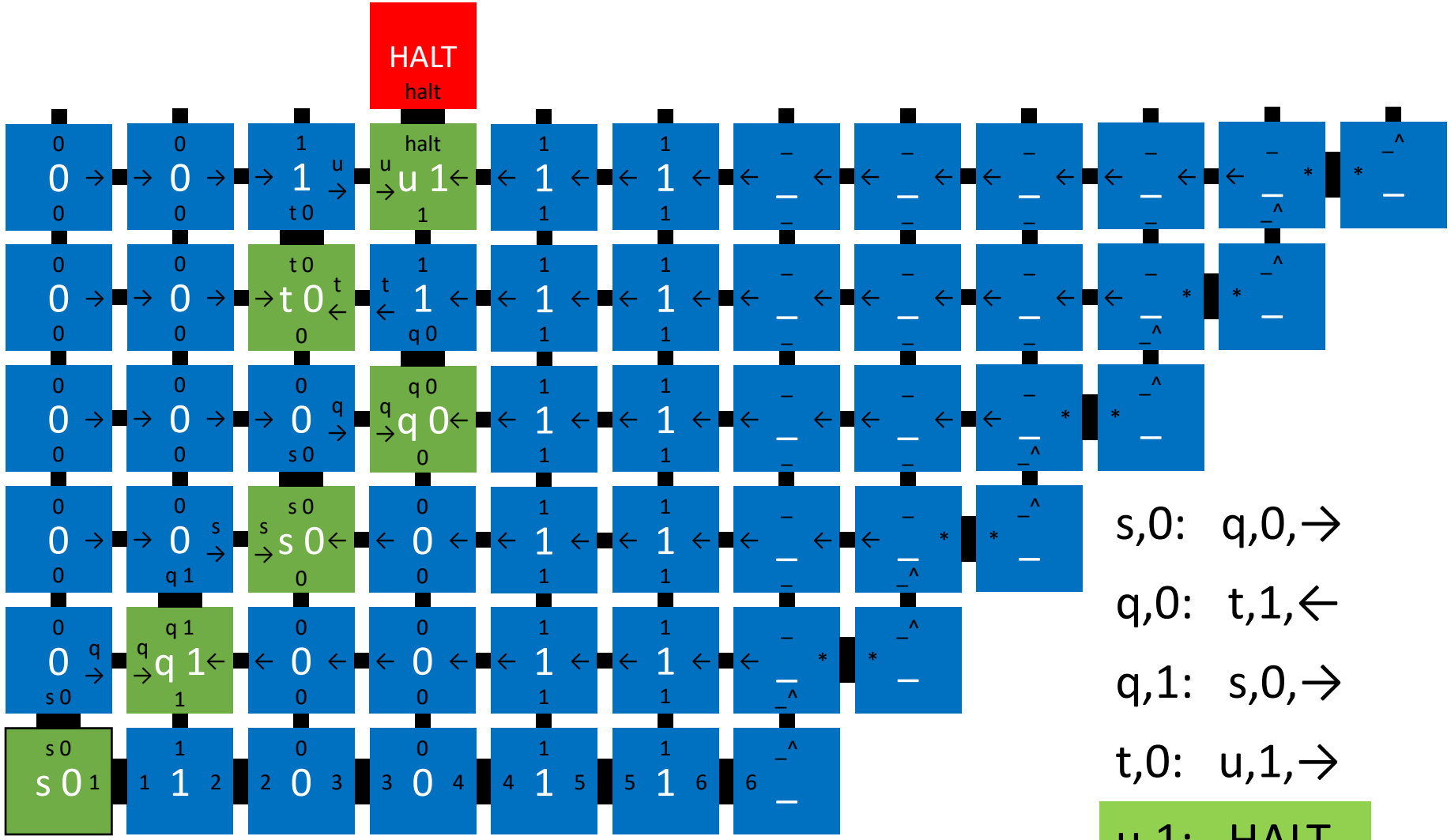

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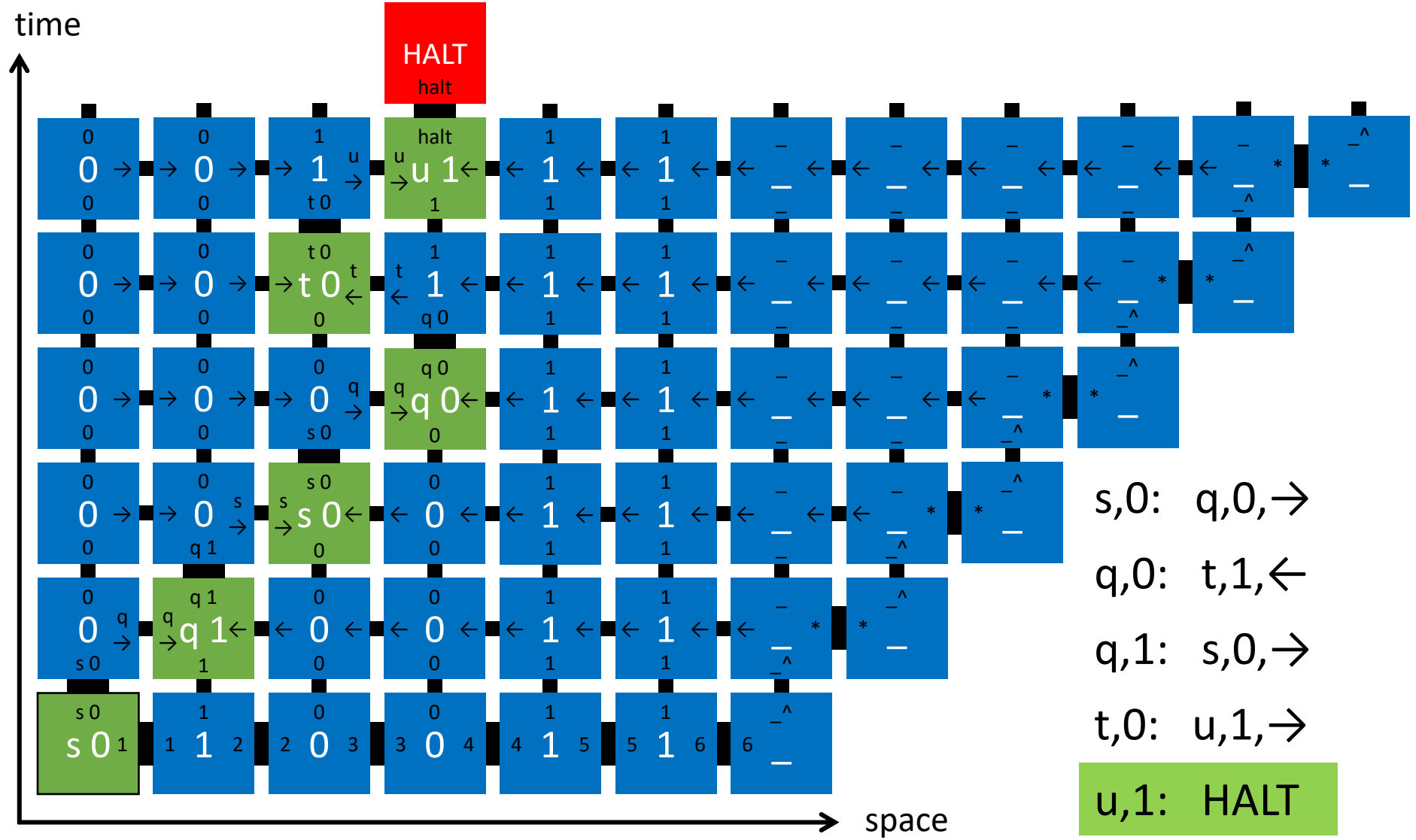

Complexity of self-assembled shapes

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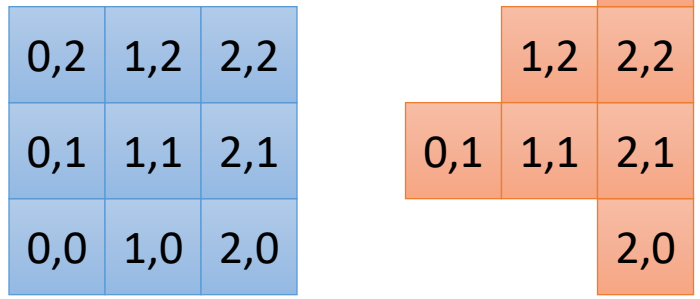

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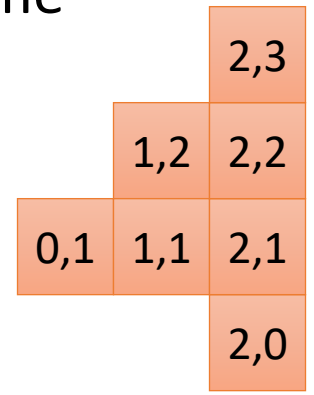

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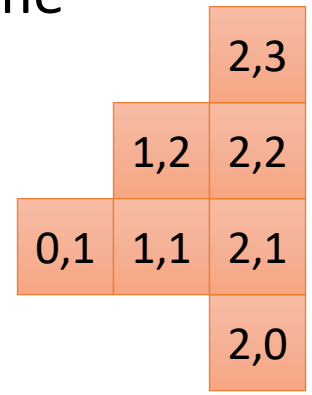

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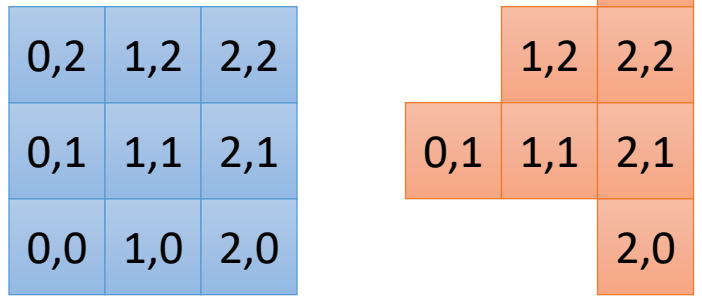

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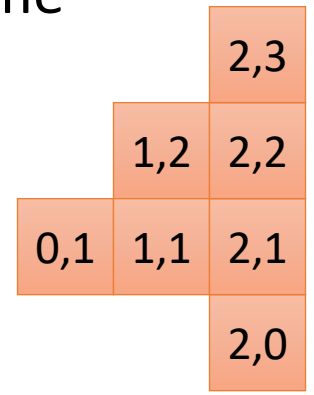

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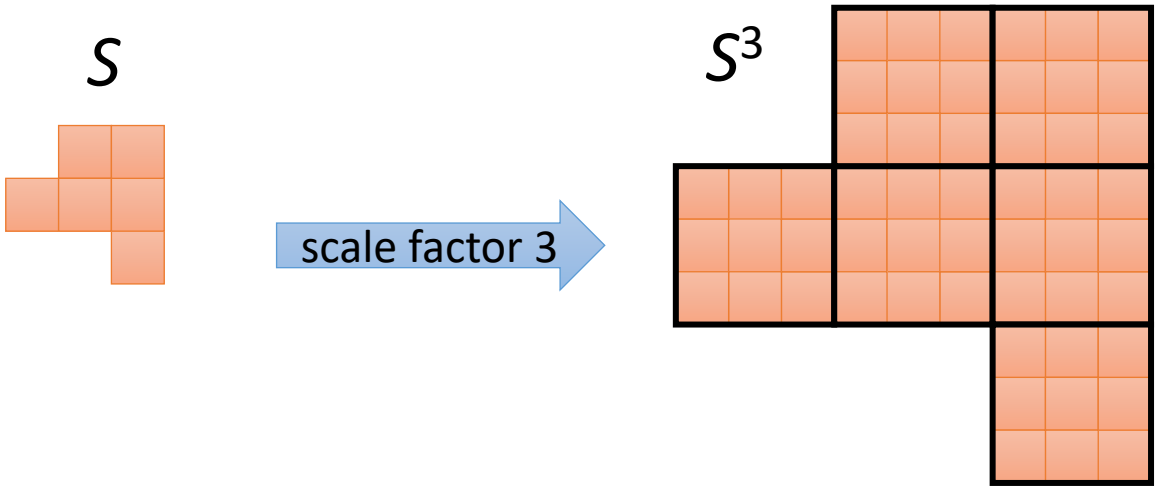

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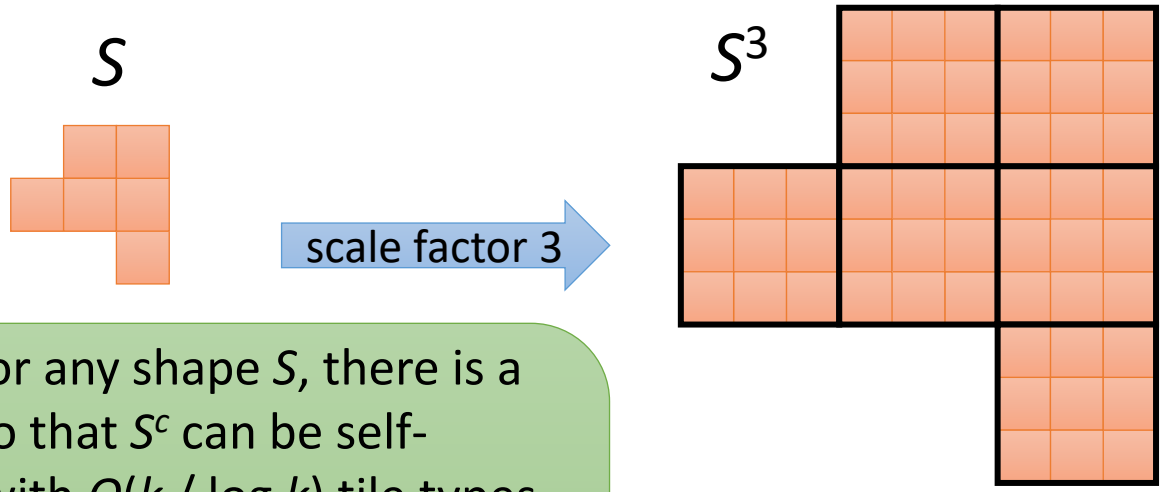

Theorem: For any shape S, there is a constant c so that S^c can be self-assembled with $O(k / \log k)$ tile types, where k is the length in bits of the shortest program (input to a universal Turing machine) that, on input (x,y), indicates whether $(x,y) \in S$.

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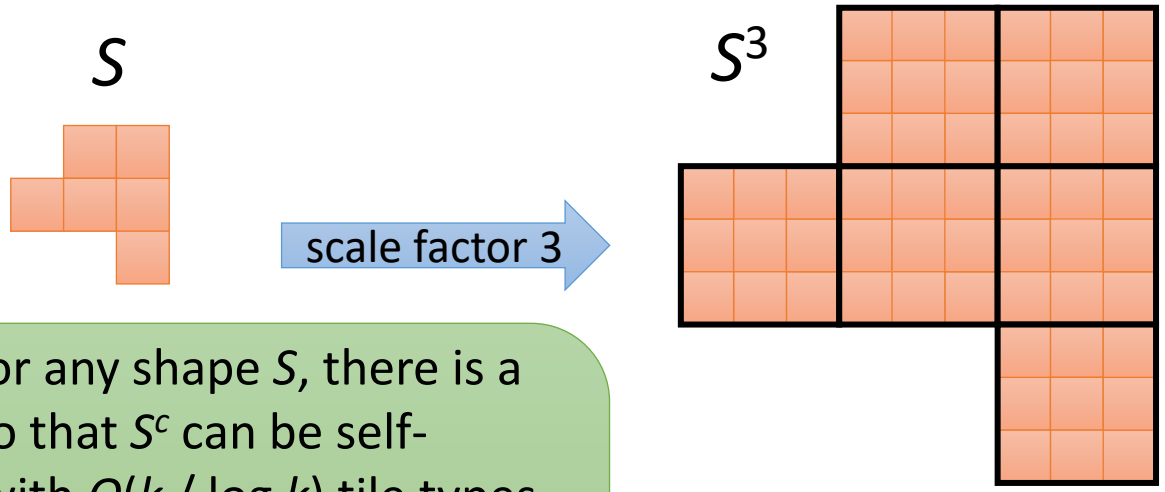

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Theorem (that we won't prove): This is optimal! No smaller tile system could self-assemble any scaling of S. If one existed, we could turn it into a program with $< k$ bits "describing" S in this way. (*Why?*)

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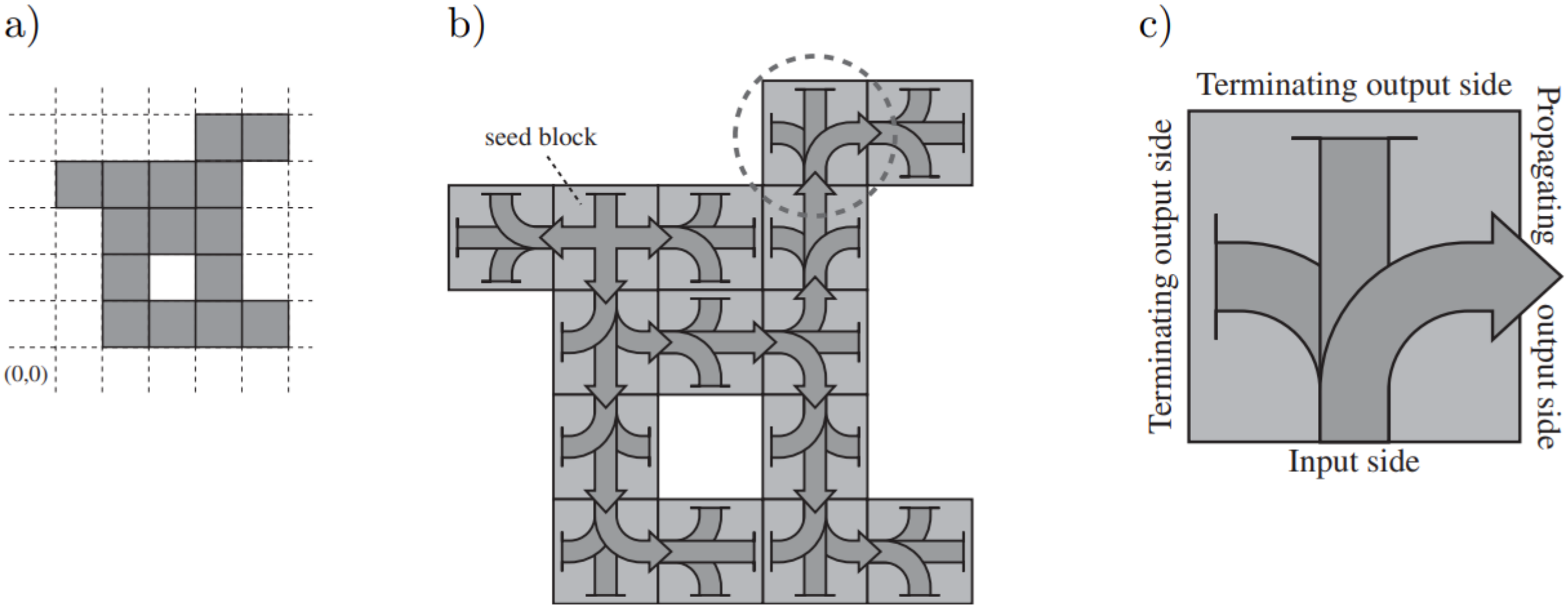

FIG. 5.1. *Forming a shape out of blocks:* (a) *A coordinated shape* S. (b) *An assembly composed of* $c \times c$ *blocks that grow according to transmitted instructions such that the shape of the final assembly is* $\tilde{S}$ *(not drawn to scale). Arrows indicate information flow and order of assembly. The seed block and the circled growth block are schematically expanded in Figure* 5.2. (c) *The nomenclature describing the types of block sides.*

Programming a shape (inaccurate cartoonish overview)

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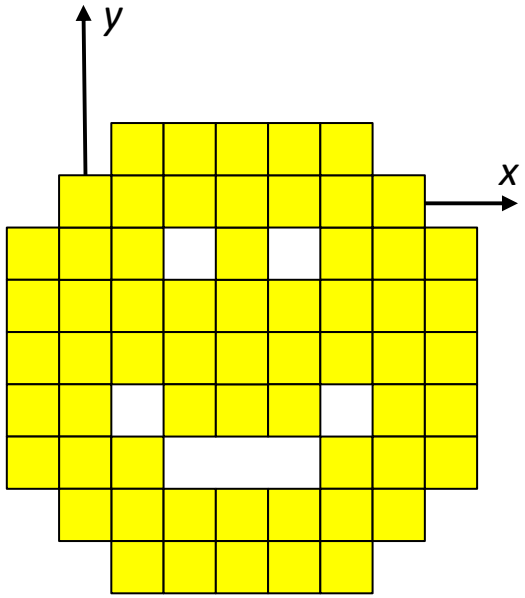

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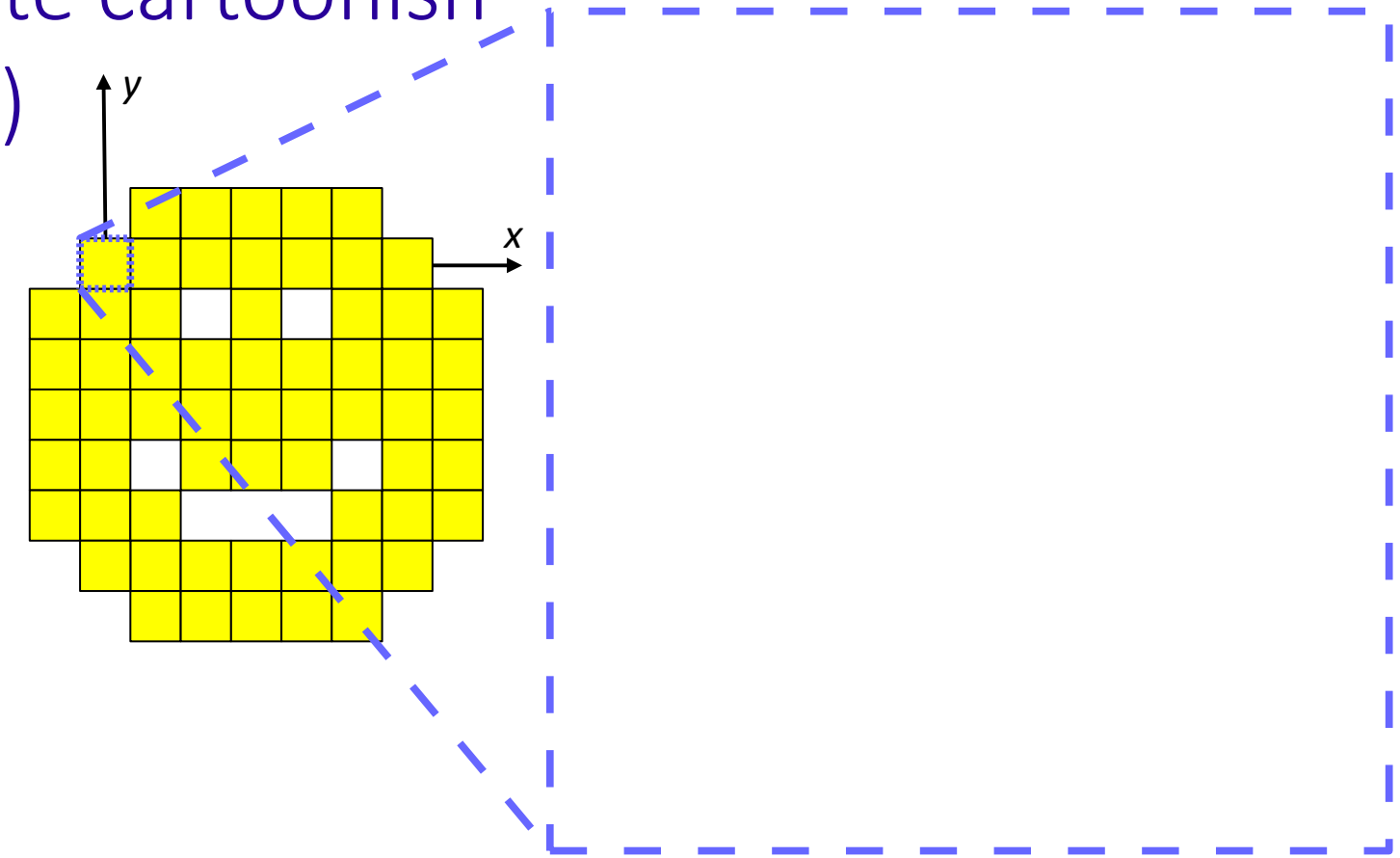

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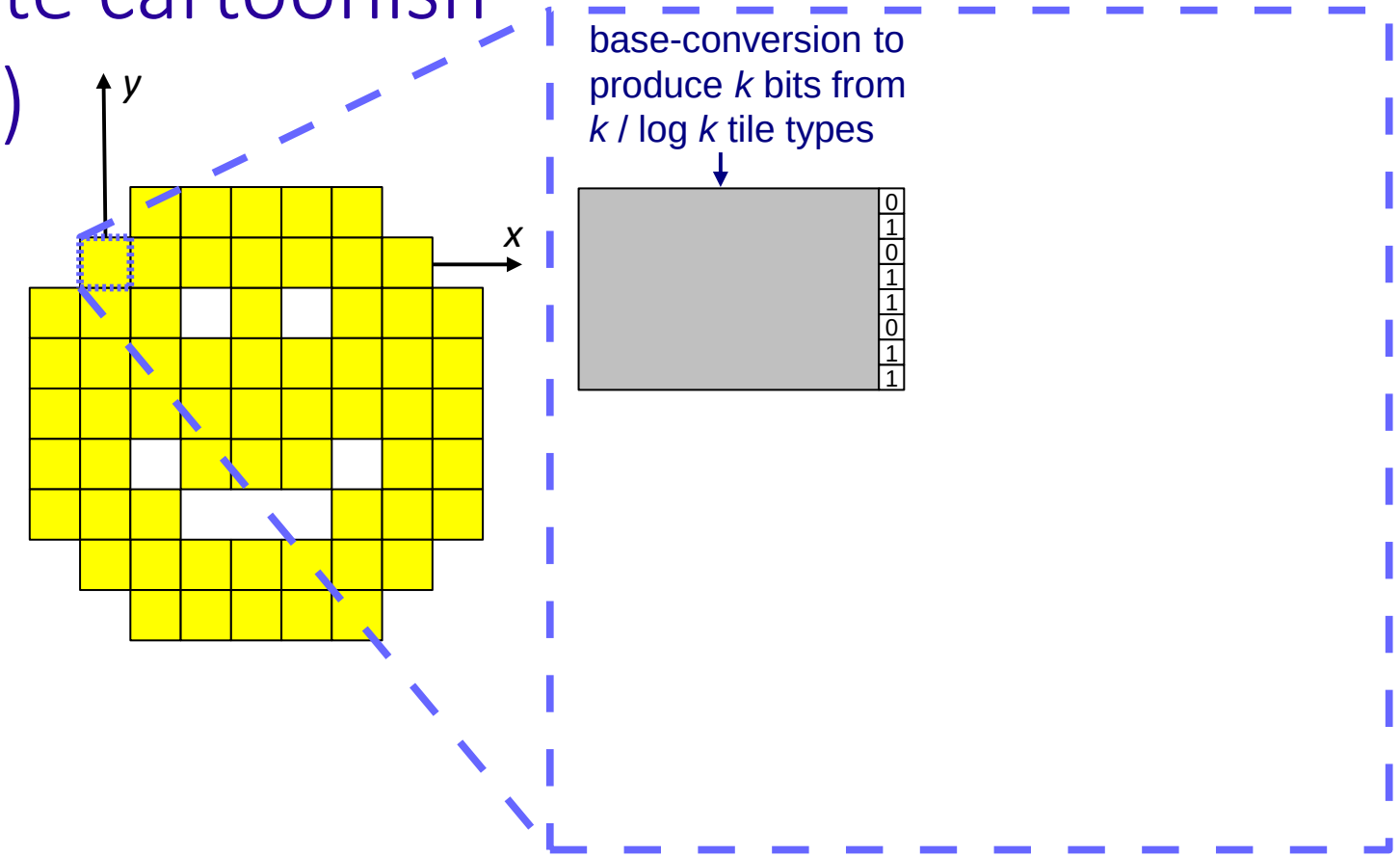

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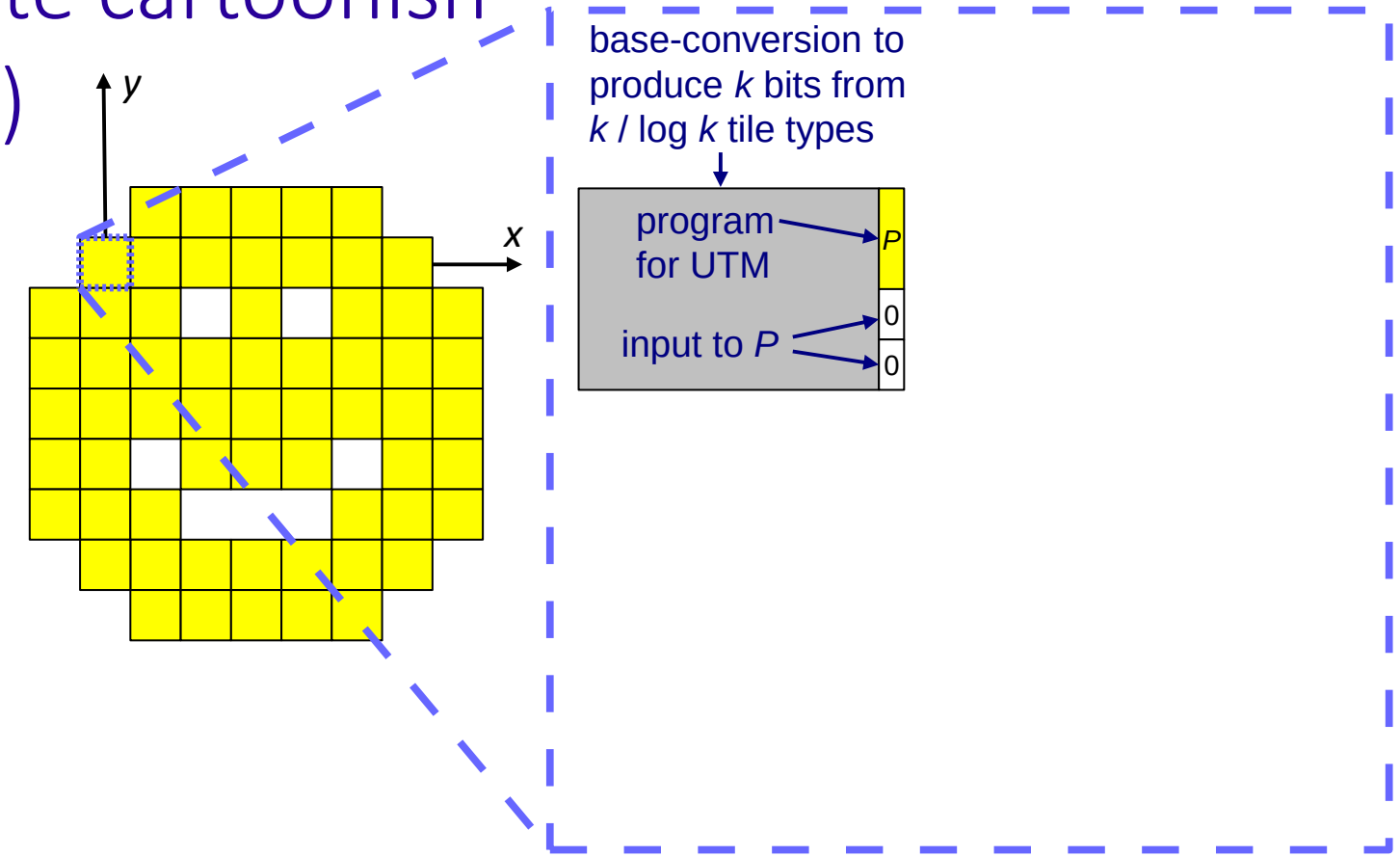

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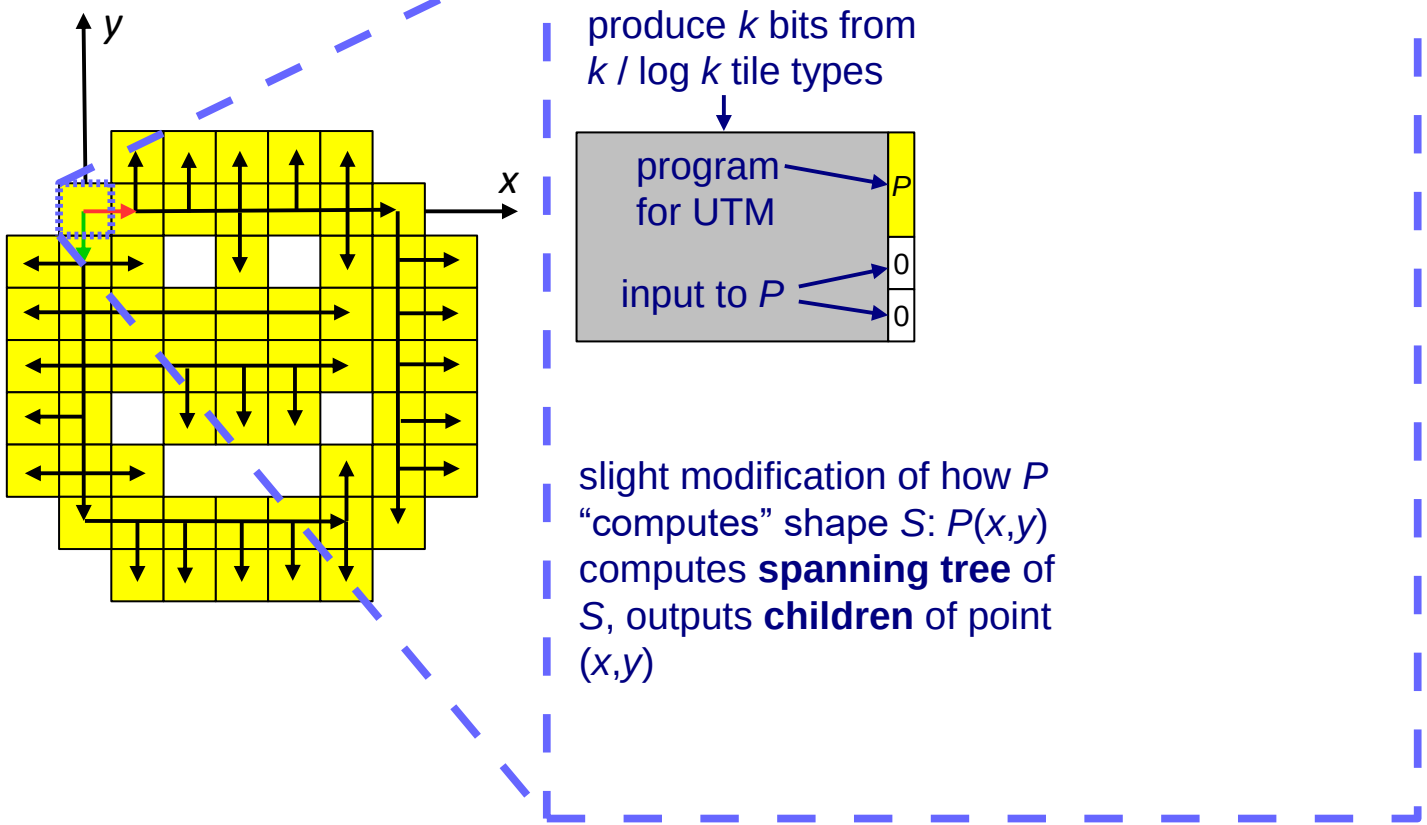

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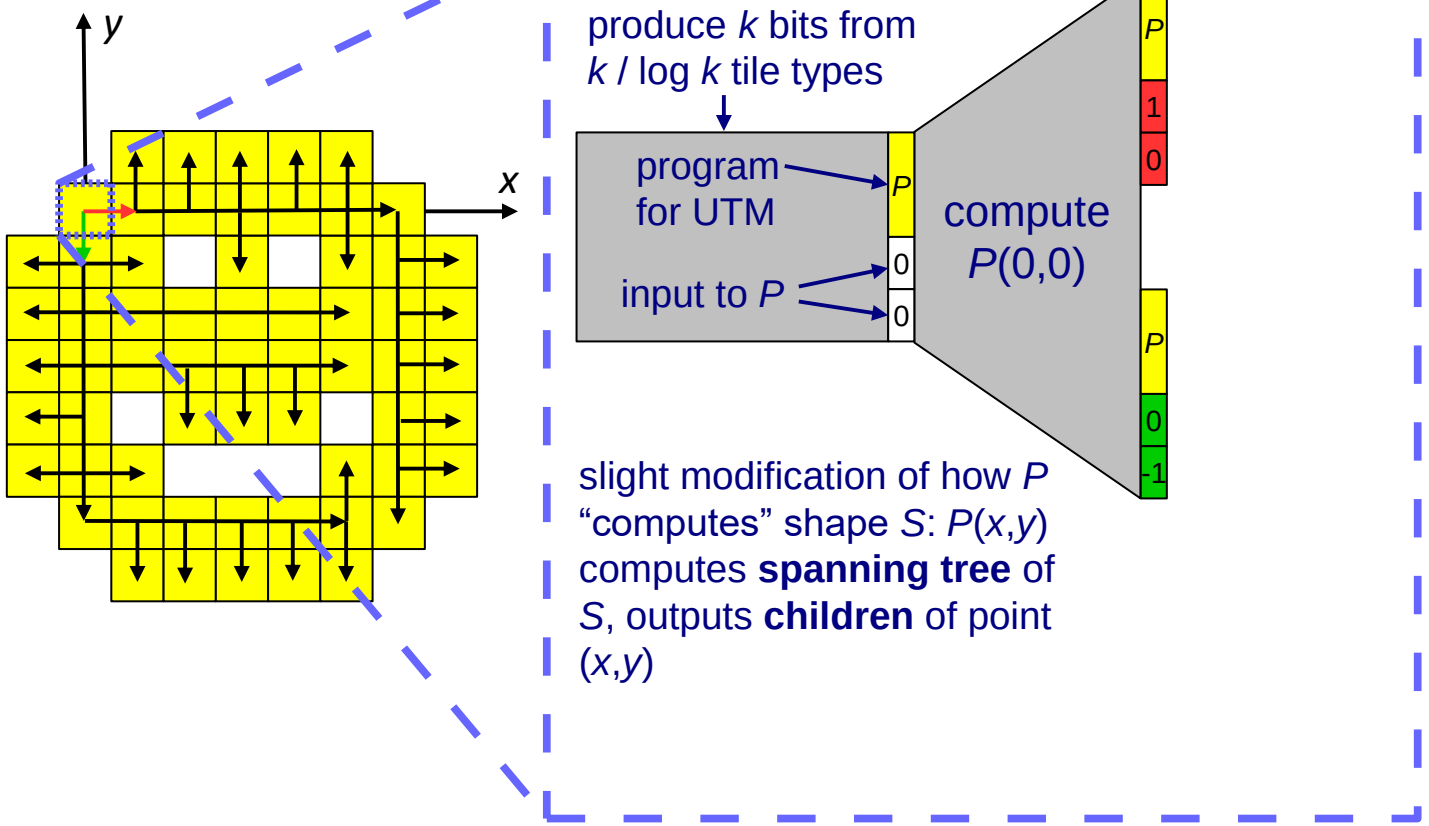

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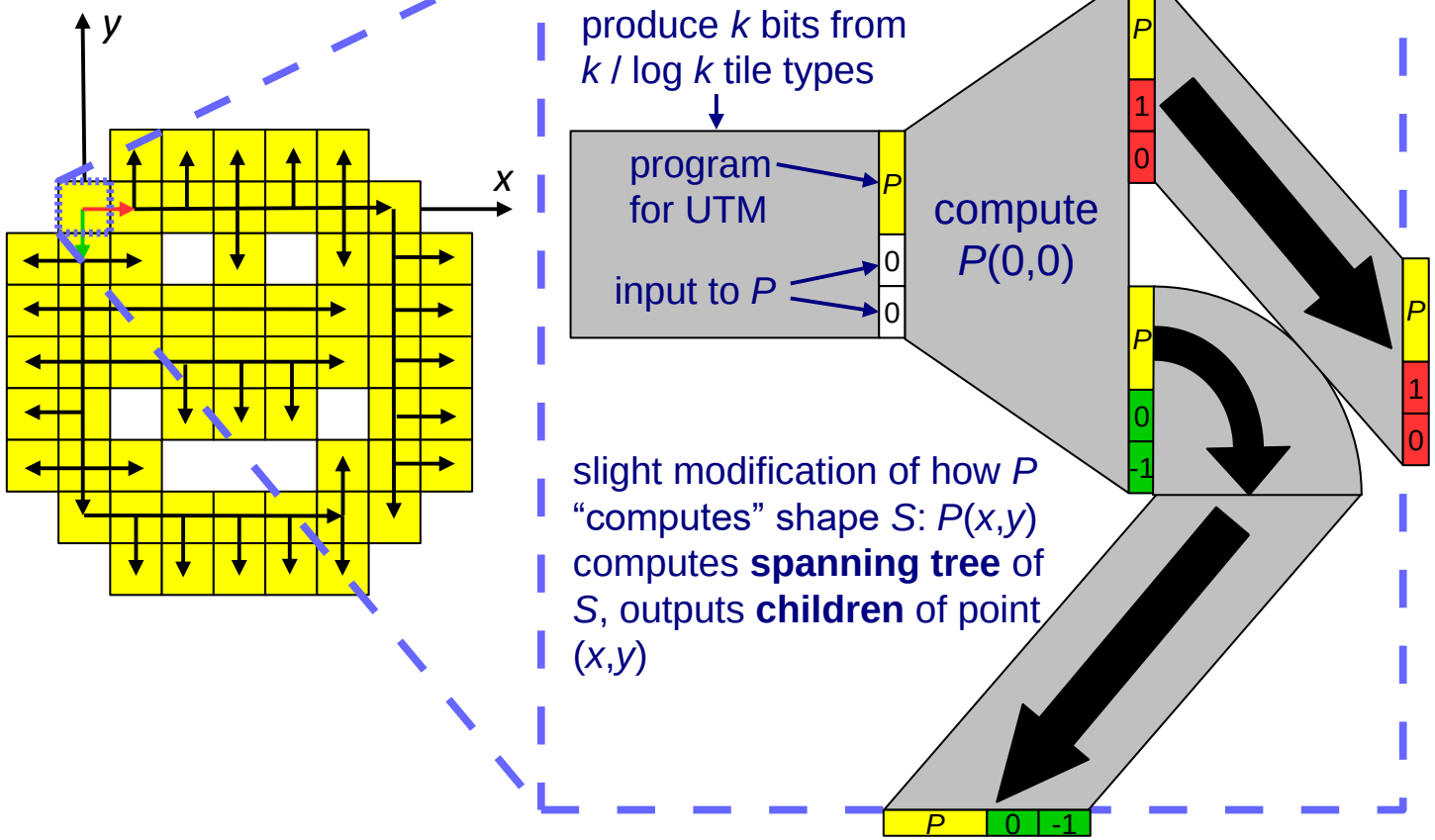

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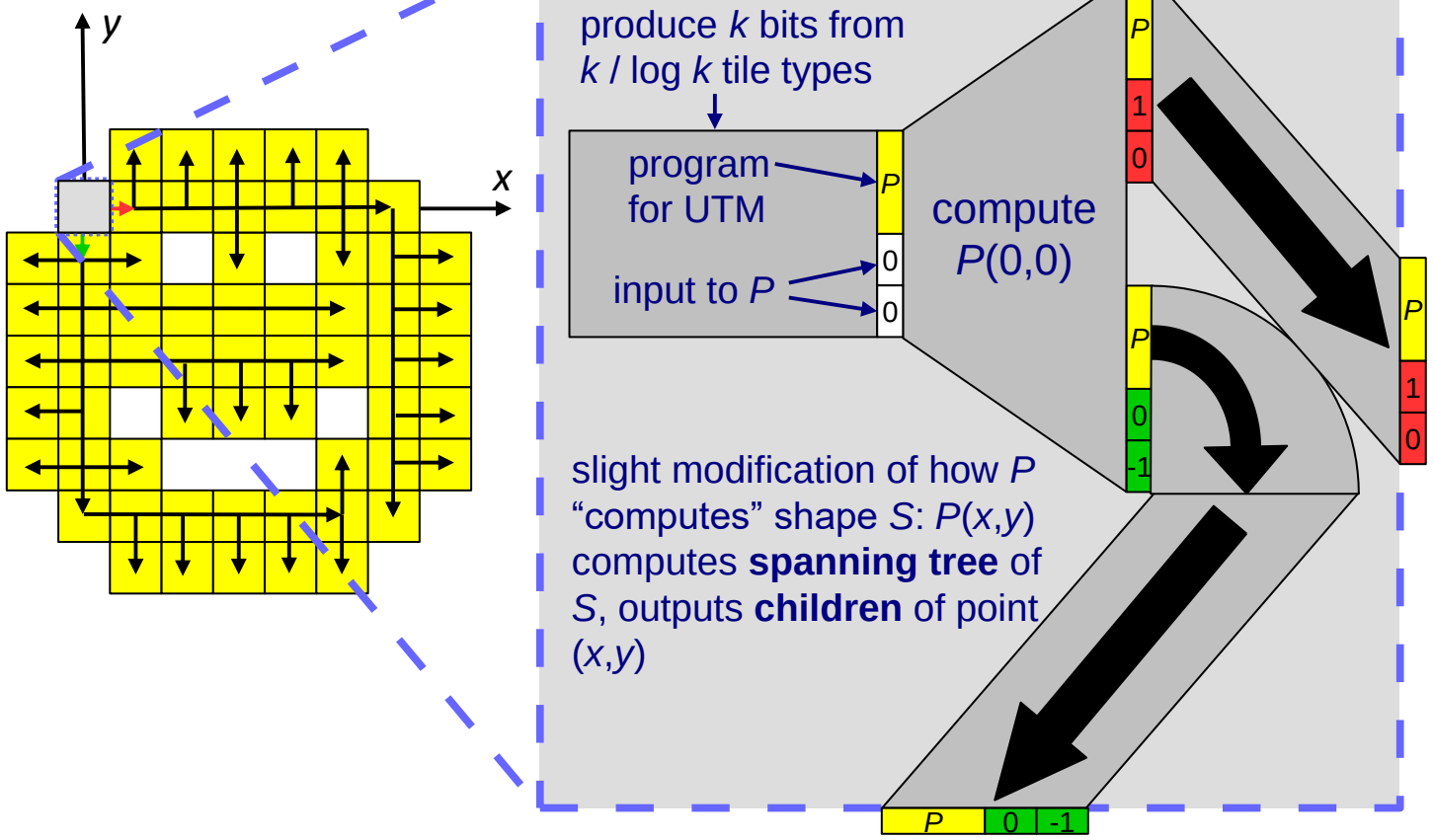

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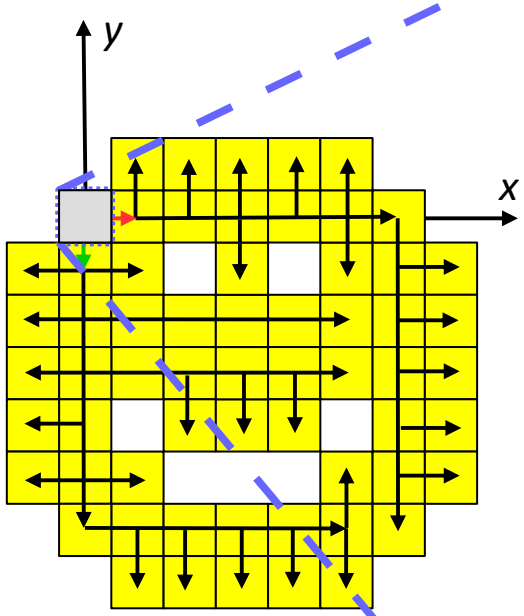

base-conversion to produce k bits from k / log k tile types

program for UTM

input to P

compute $P(0,0)$

slight modification of how P "computes" shape S: $P(x,y)$ computes **spanning tree** of S, outputs **children** of point (x,y)

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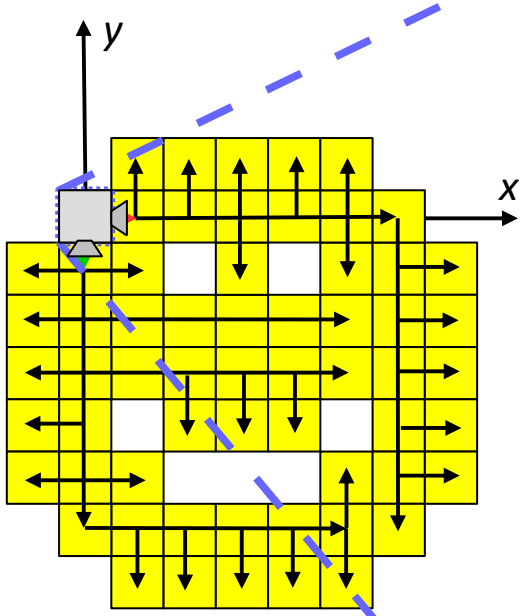

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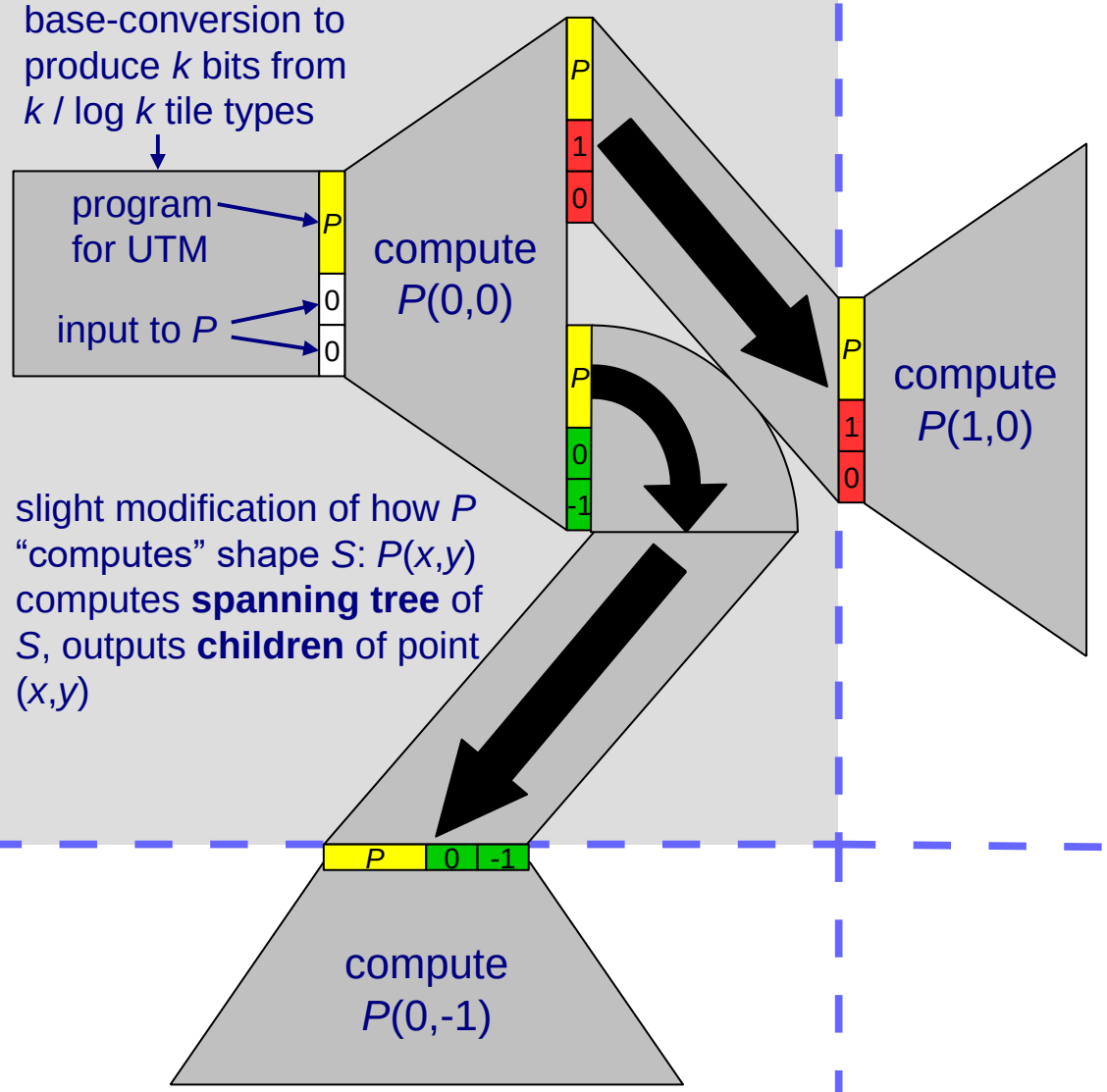

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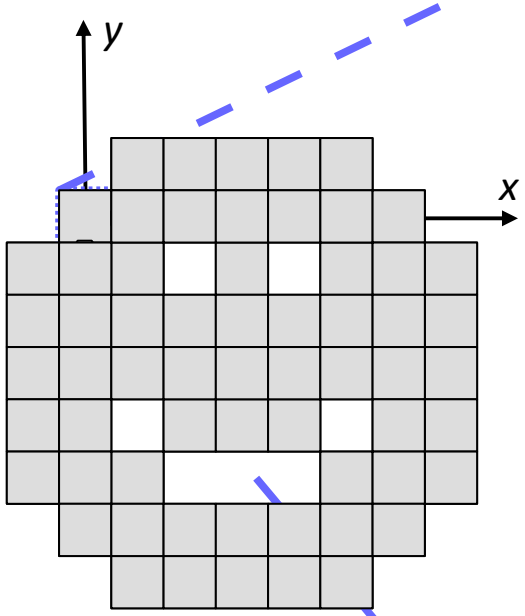

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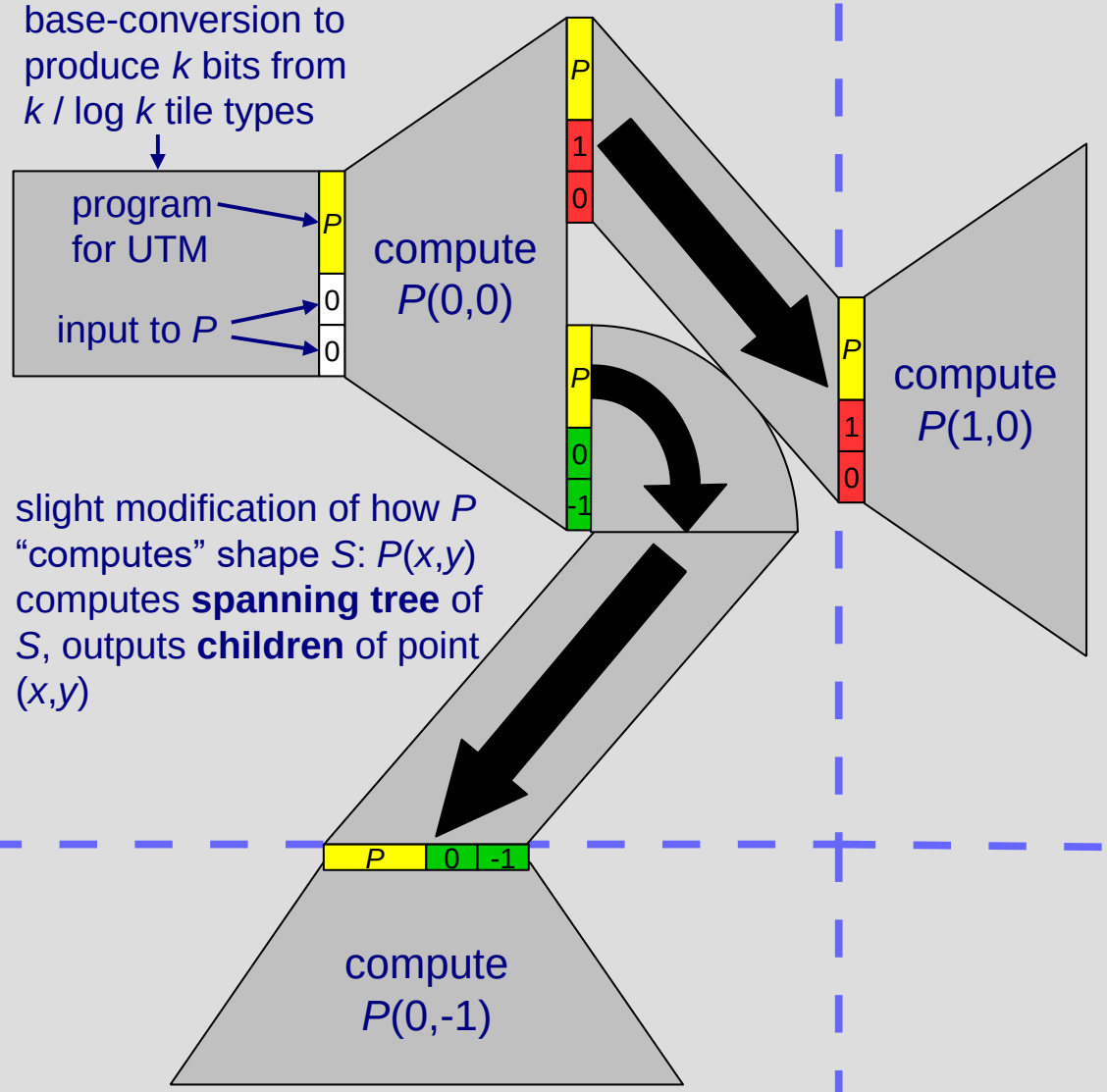

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More accurate detailed overview

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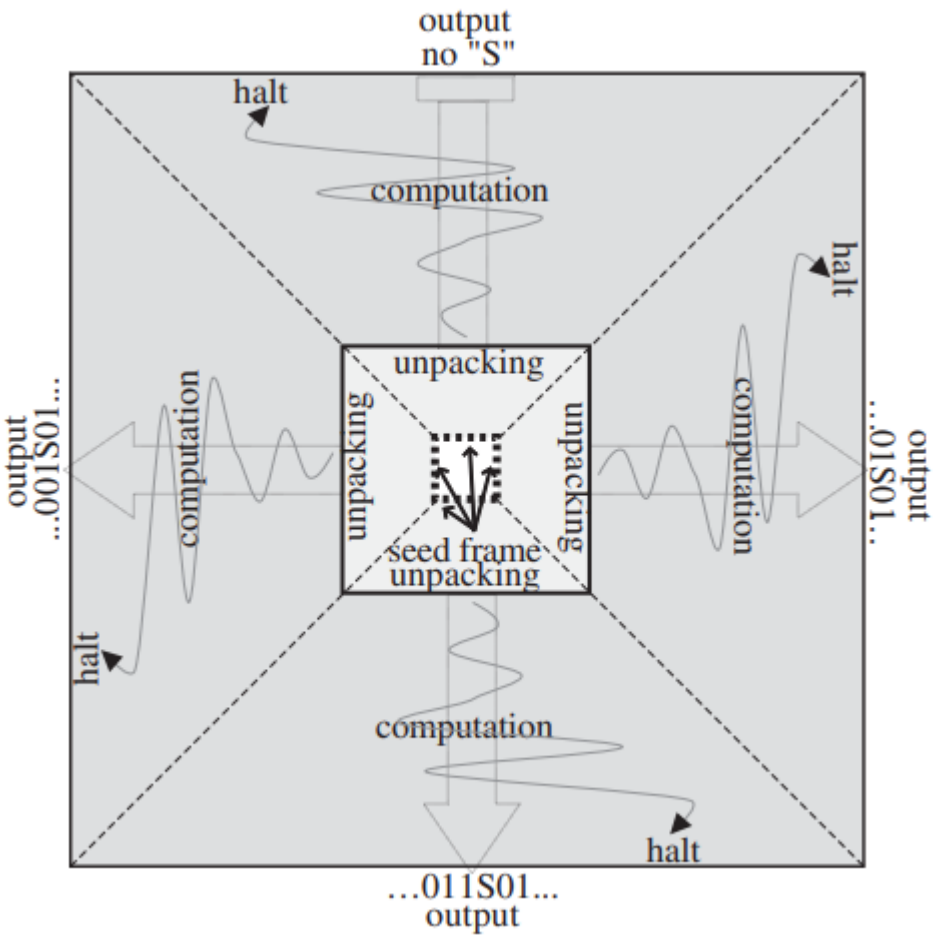
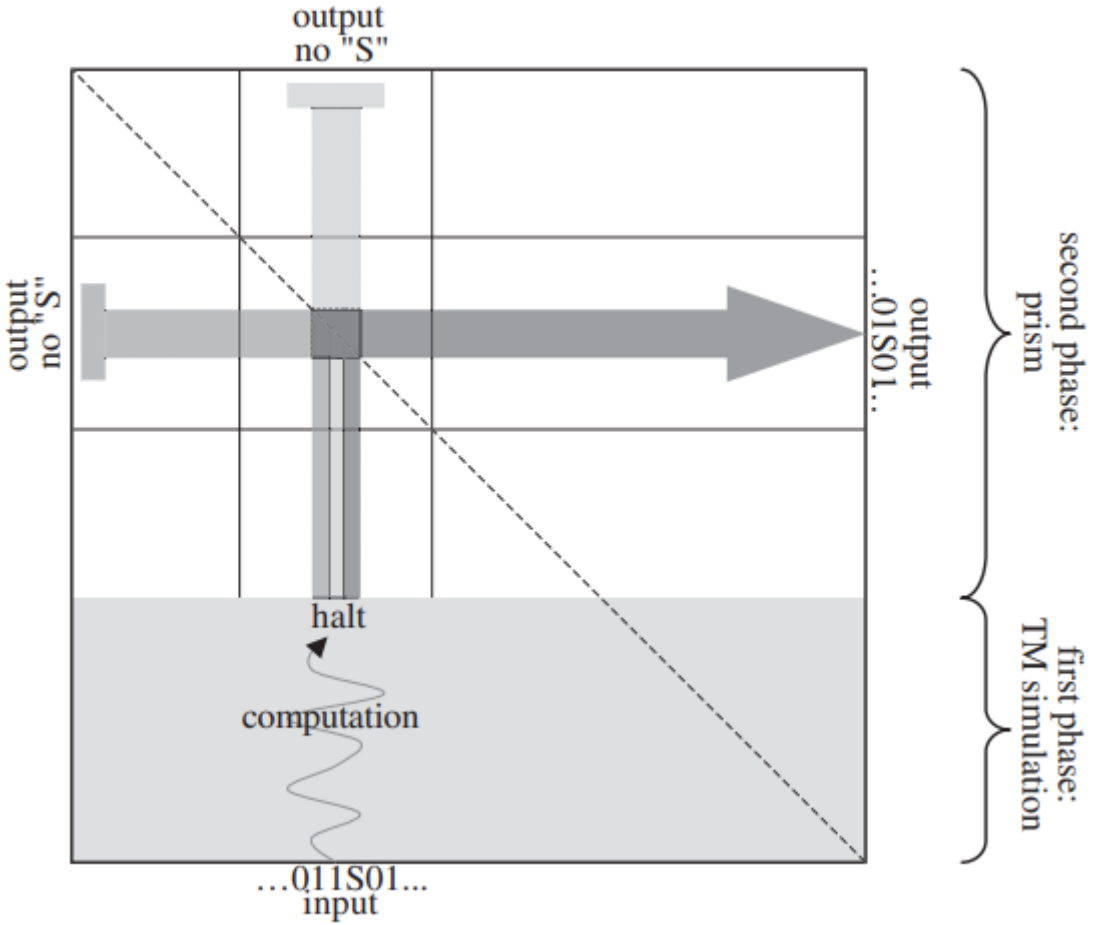
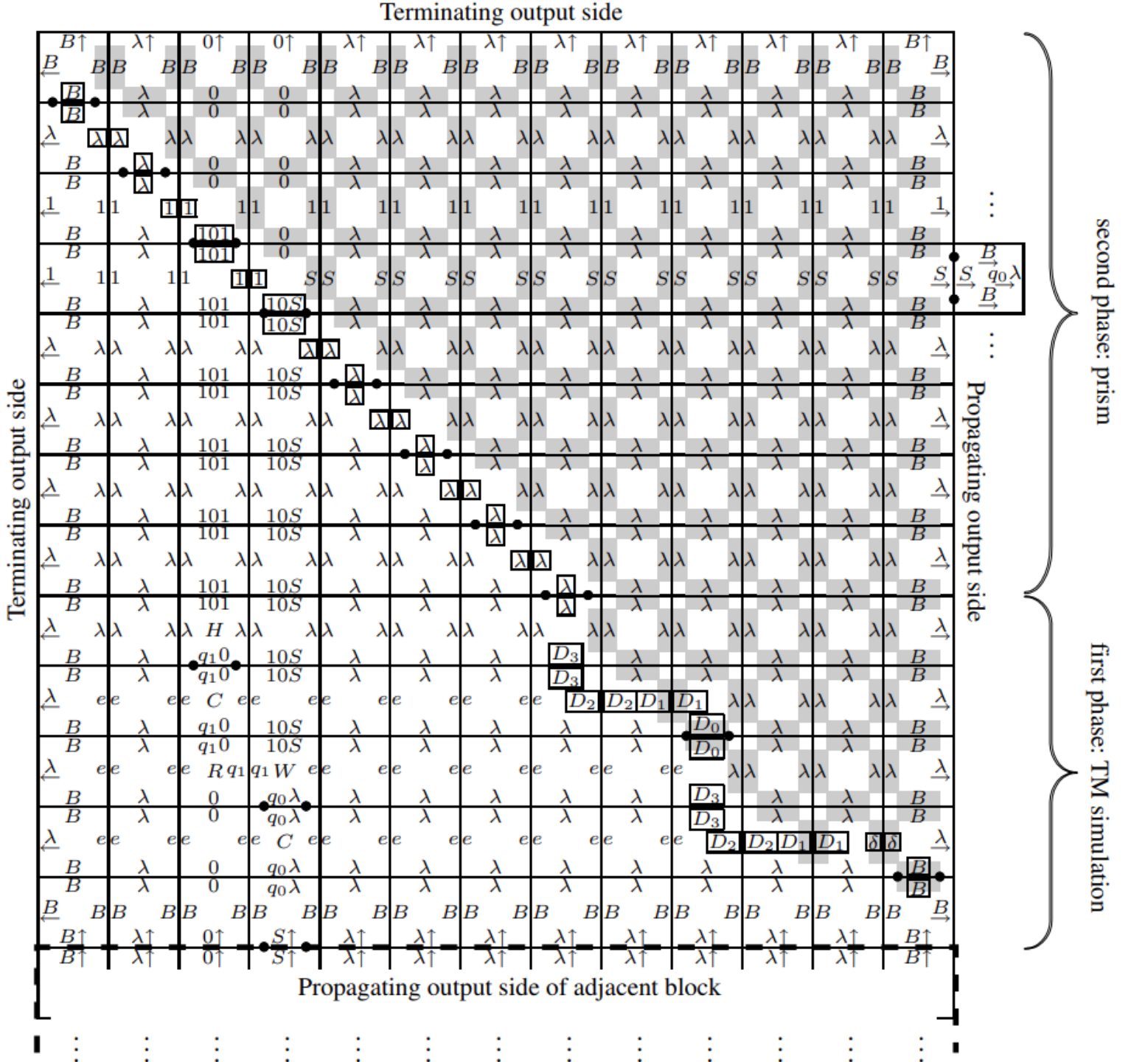


growth block



fully-detailed example of growth block



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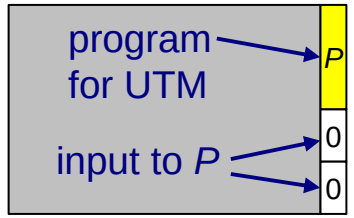

Two interpretations

i.e., T is a **universal** set of tile types that can self-assemble any shape, by giving it the right seed.

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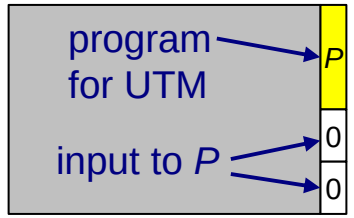

Strict and weak self-assembly

Computability-theoretic questions about self-assembly

Strict and weak self-assembly

Recall:

Let $X \subseteq \mathbb{Z}^2$ be a **shape**, a connected subset of $\mathbb{Z}^2$. Θ **strictly self-assembles** X if, for all $\alpha \in A_{\square}[\Theta]$, $S_\alpha = X$.
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Let $X \subseteq \mathbb{Z}^2$. Θ **weakly self-assembles** X if there is a subset $B \subseteq T$ (the "blue tiles") such that, for all $\alpha \in A_{\square}[\Theta]$, $X = \alpha^{-1}(B)$.
(*every terminal producible assembly puts blue tiles exactly on X.*)

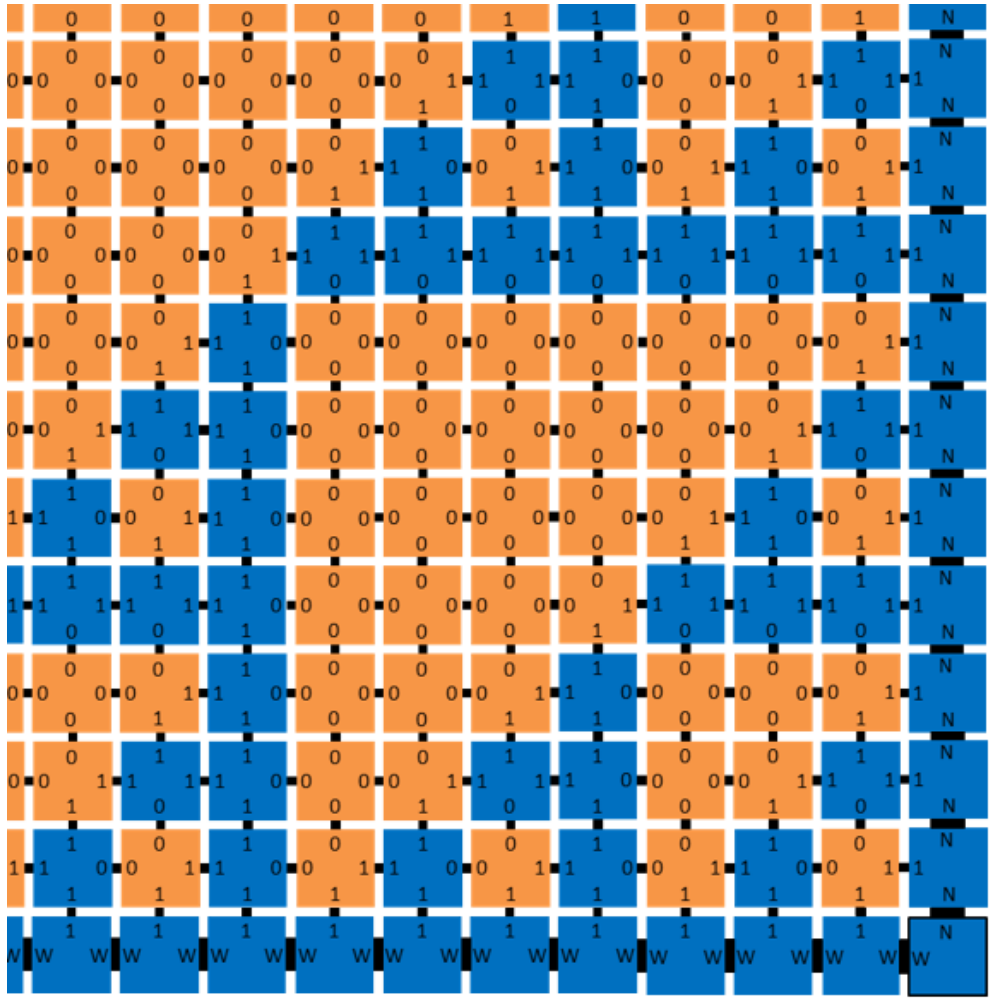
Strict and weak self-assembly

Recall:

Let $X \subseteq \mathbb{Z}^2$ be a **shape**, a connected subset of $\mathbb{Z}^2$. Θ **strictly self-assembles** X if, for all $\alpha \in A_{\square}[\Theta]$, $S_\alpha = X$.
(*every terminal producible assembly has shape X*)

Let $X \subseteq \mathbb{Z}^2$. Θ **weakly self-assembles** X if there is a subset $B \subseteq T$ (the "blue tiles") such that, for all $\alpha \in A_{\square}[\Theta]$, $X = \alpha^{-1}(B)$.
(*every terminal producible assembly puts blue tiles exactly on X.*)

Tile system on right strictly self-assembles the whole second quadrant, and it weakly self-assembles the discrete Sierpinski triangle.



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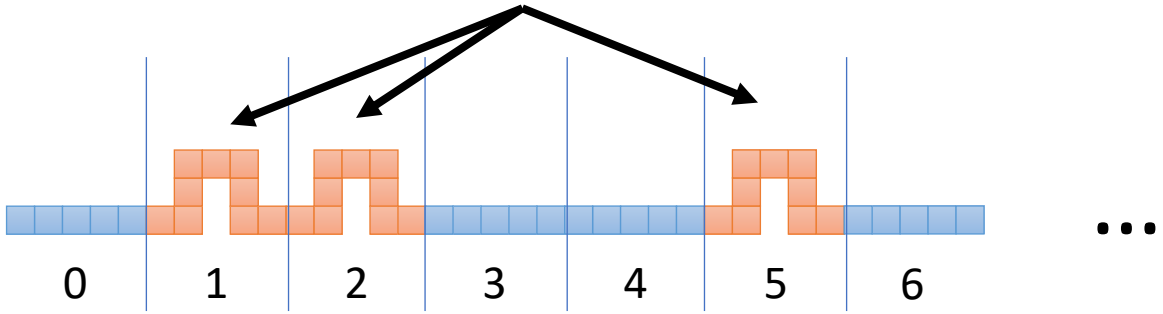

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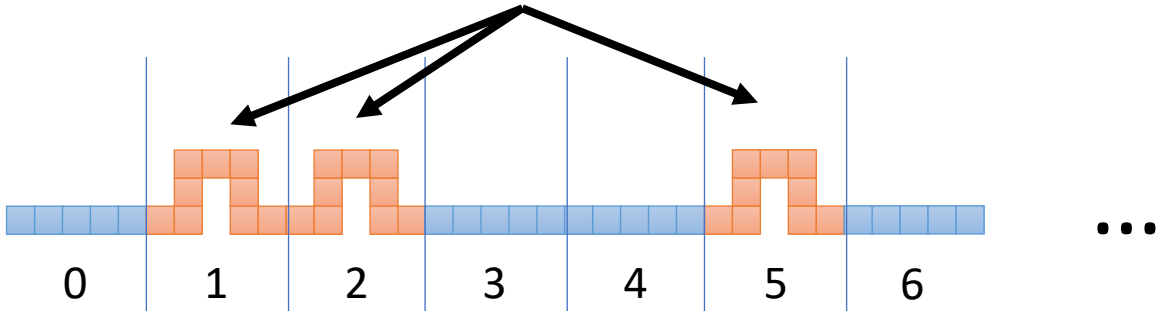

Question: Is there a computable shape $S \subseteq \mathbb{Z}^2$ that cannot be strictly self-assembled?

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- Let V = { (0,0), (0,1), (1,0) } be three vectors for “recursive translation”.

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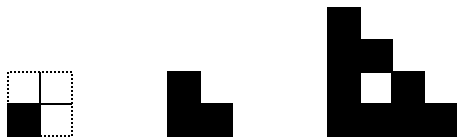

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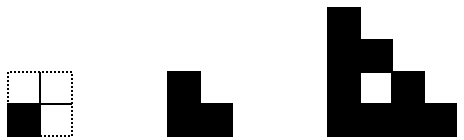

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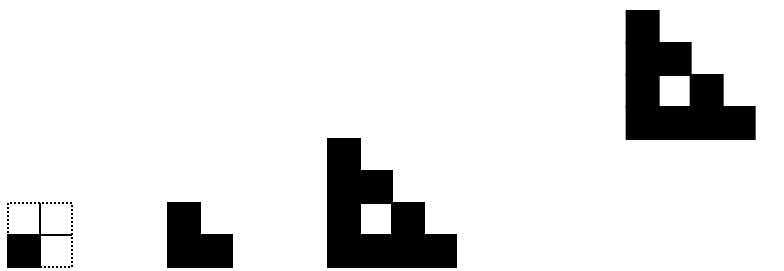

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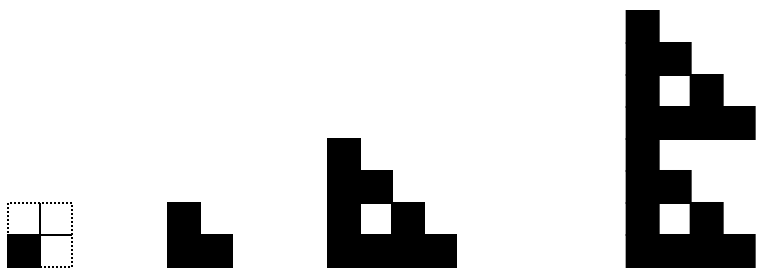

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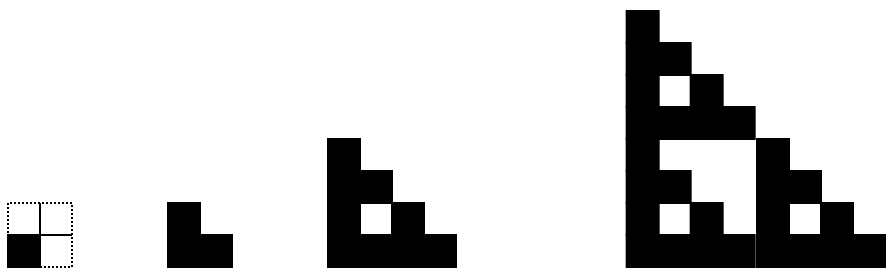

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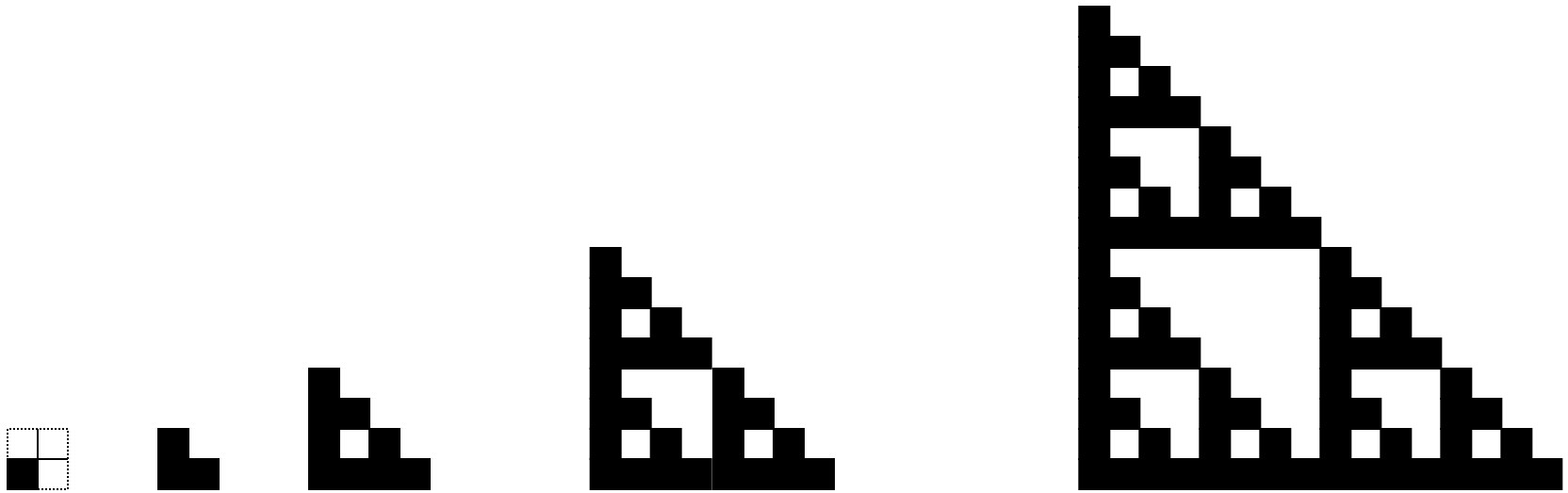

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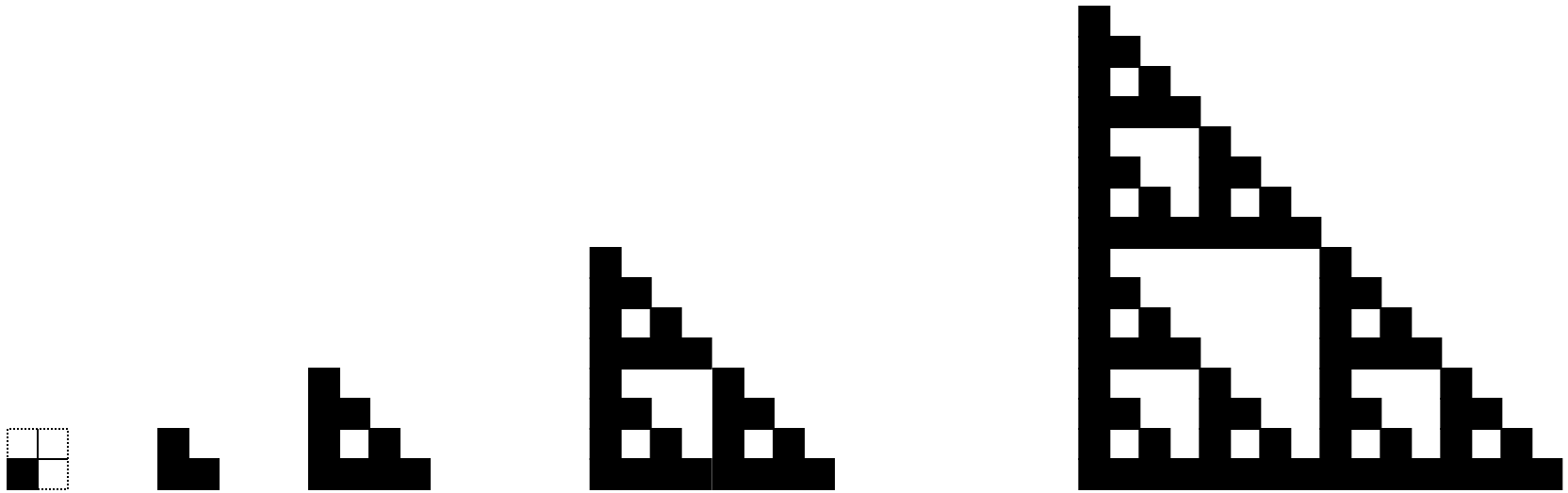

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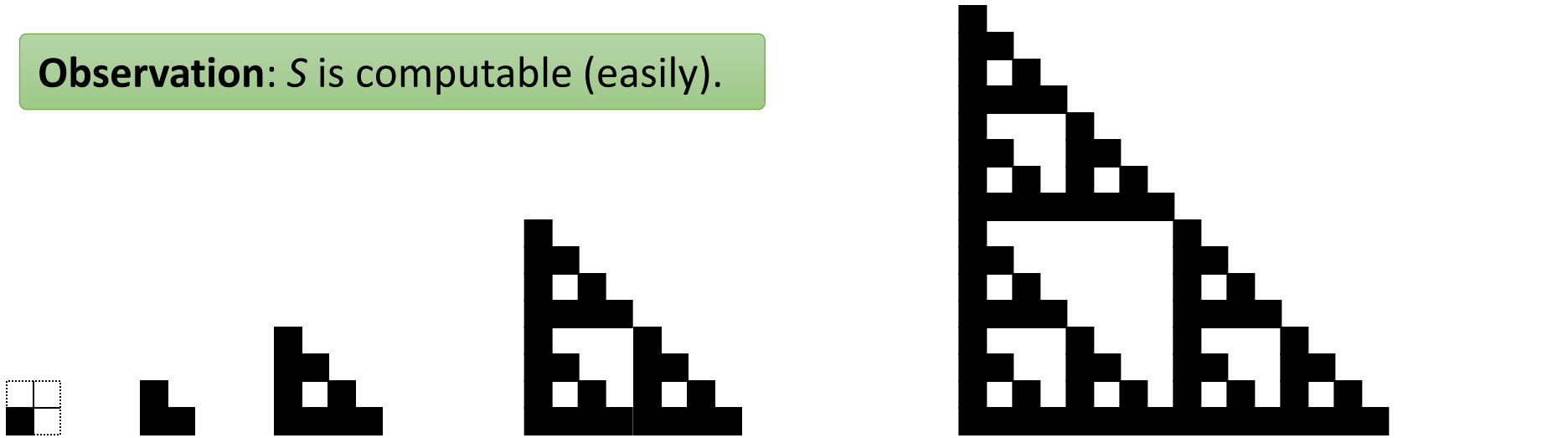
Observation: *S* is computable (easily).



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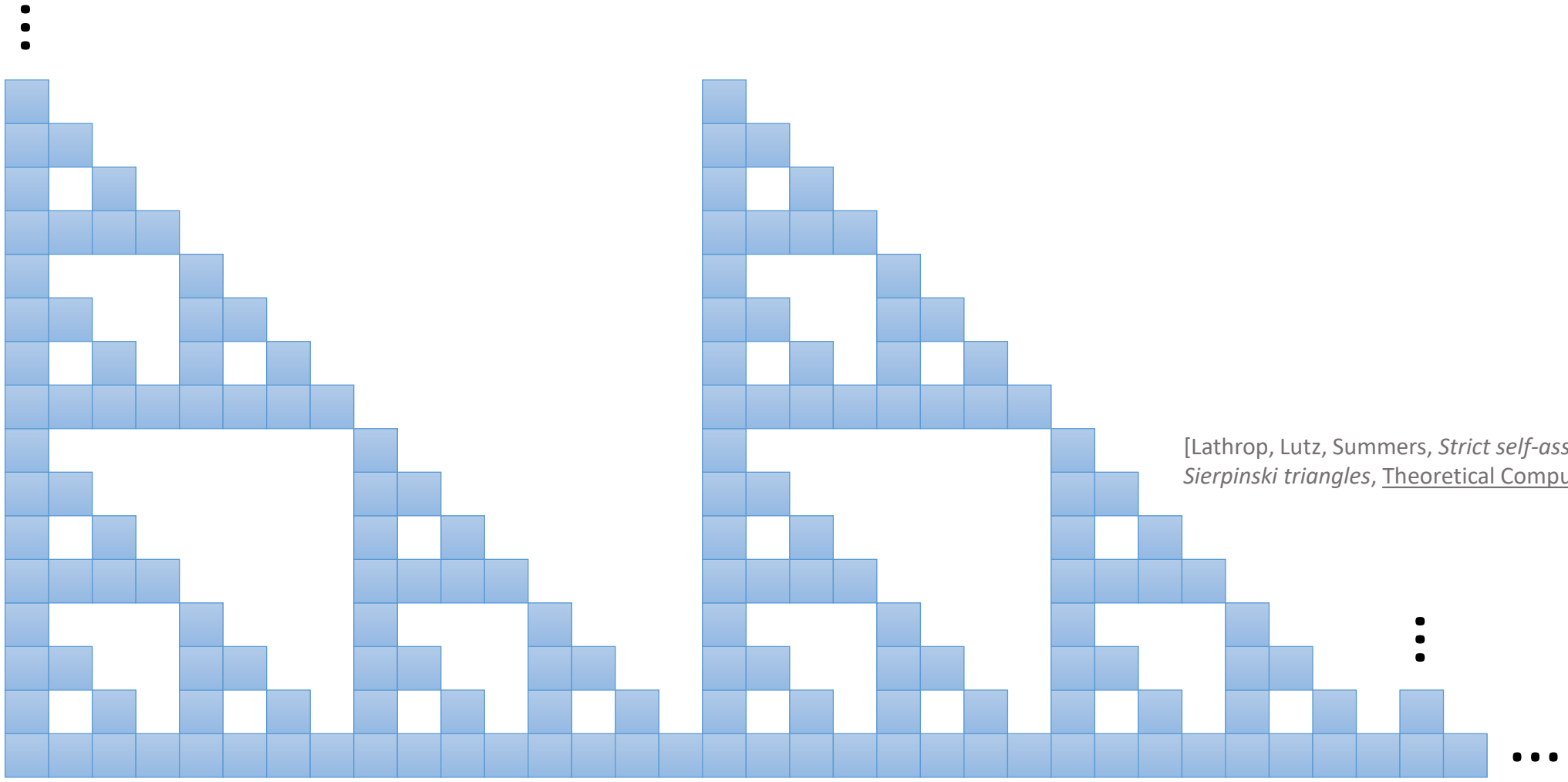

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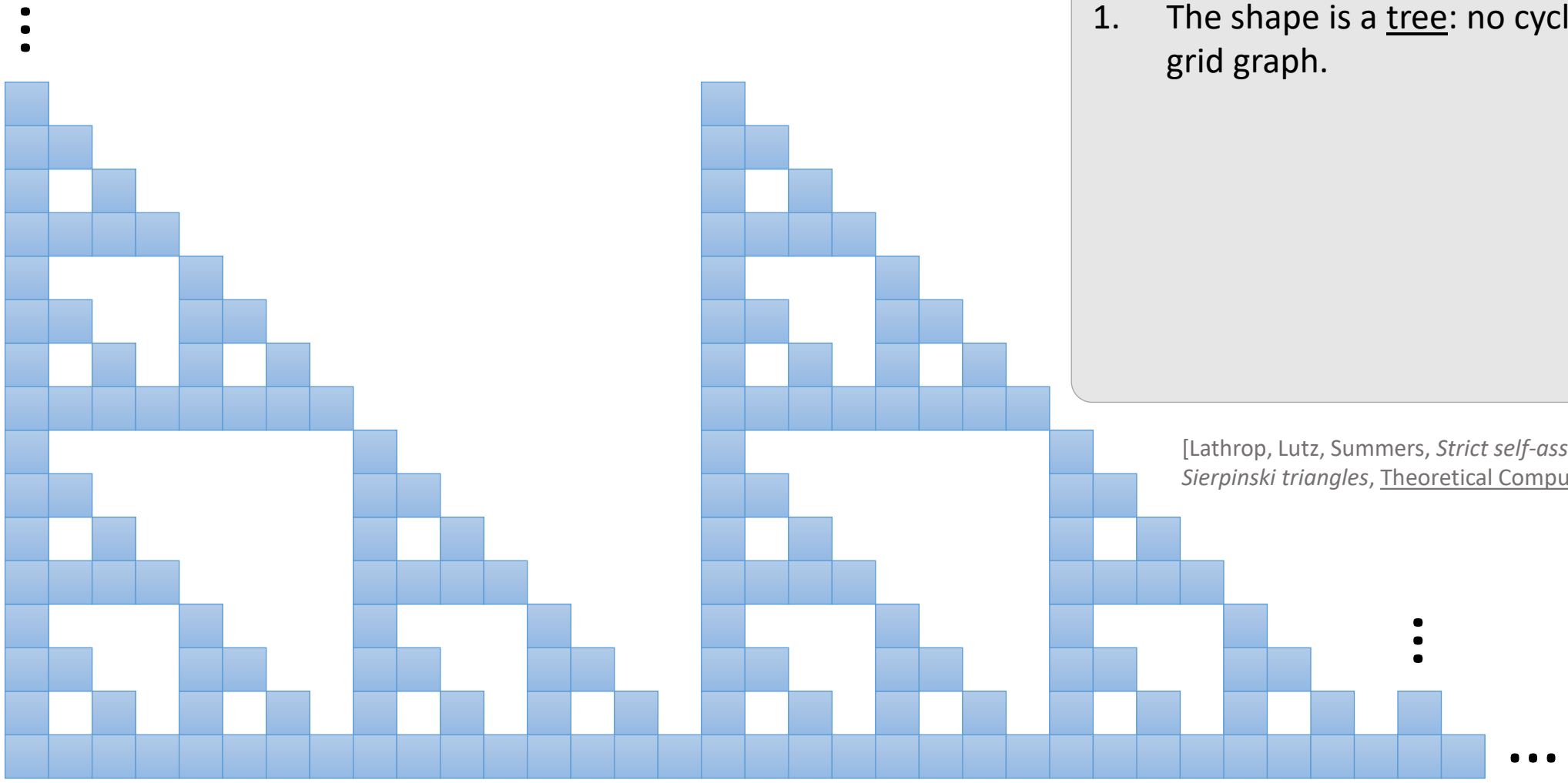

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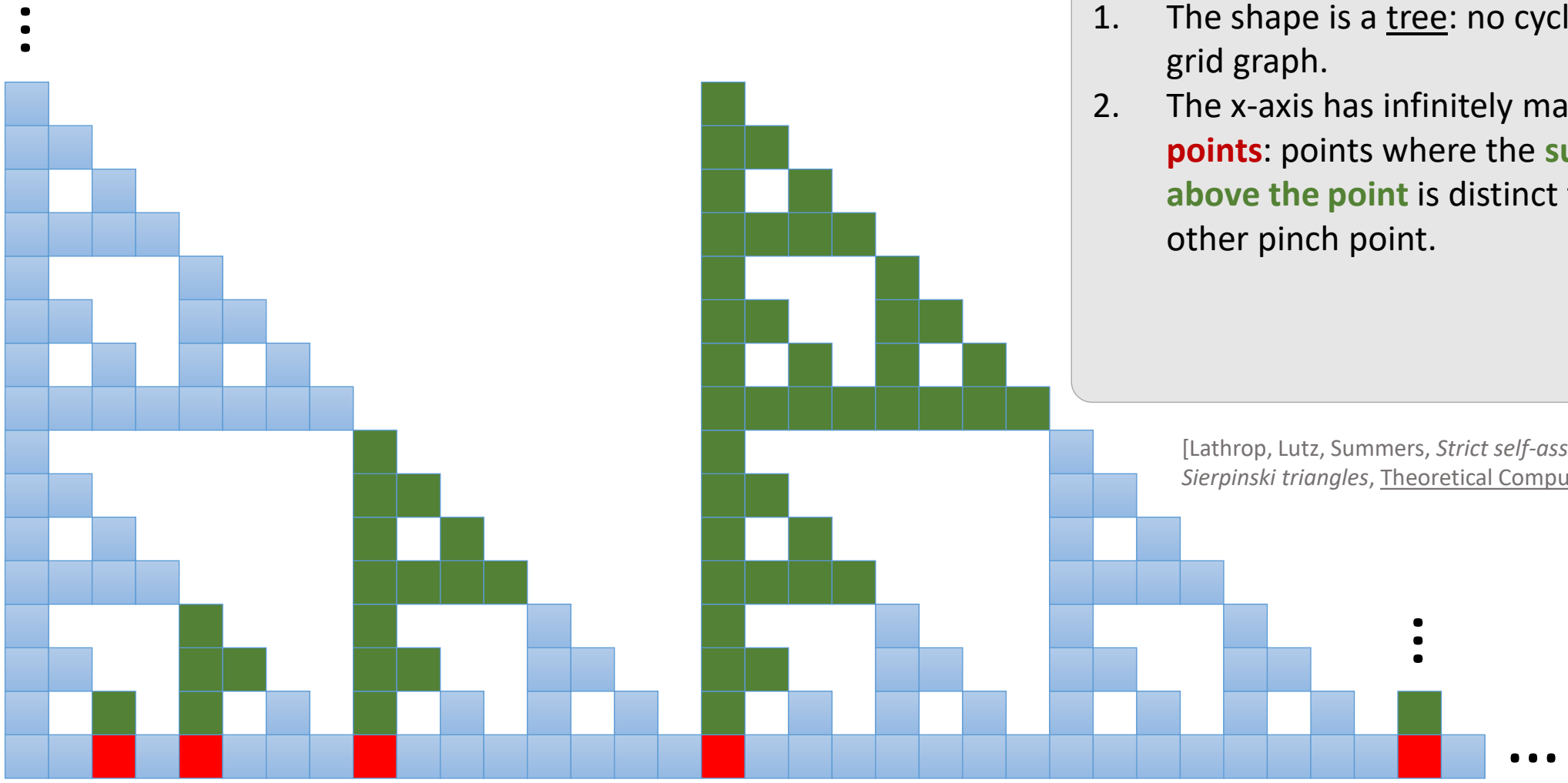

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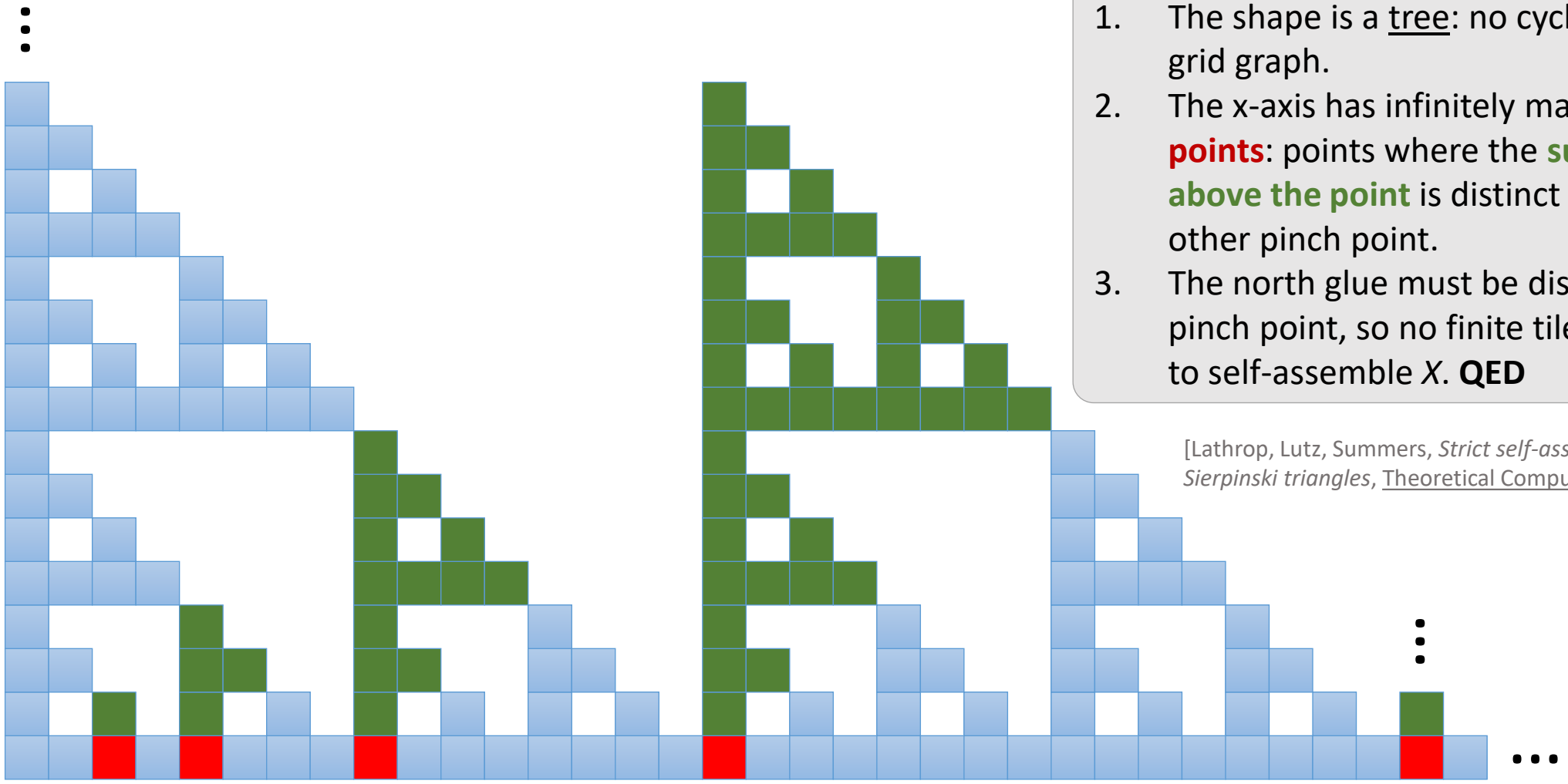

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3. The north glue must be distinct at each pinch point, so no finite tile set suffices to self-assemble *X*. **QED**

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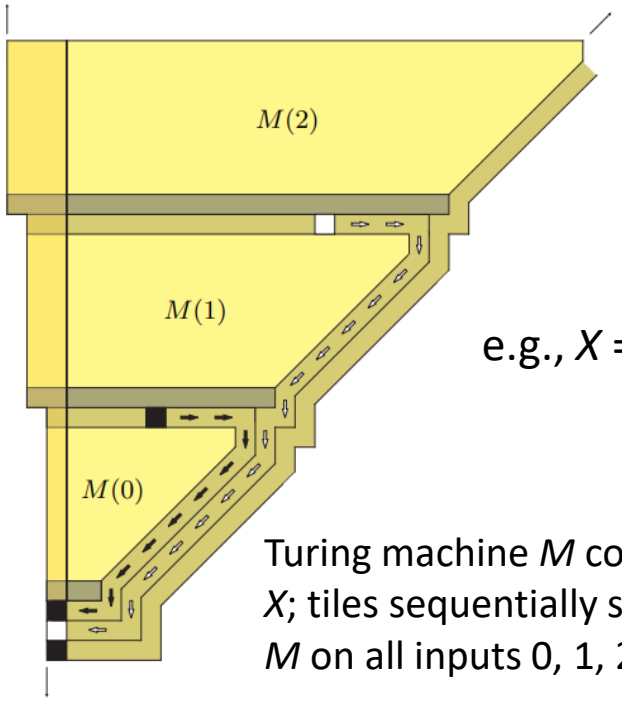
Theorem: Every computable set $X \subseteq \mathbb{N}$, "embedded straightforwardly" in $\mathbb{Z}^2$, can be weakly self-assembled.



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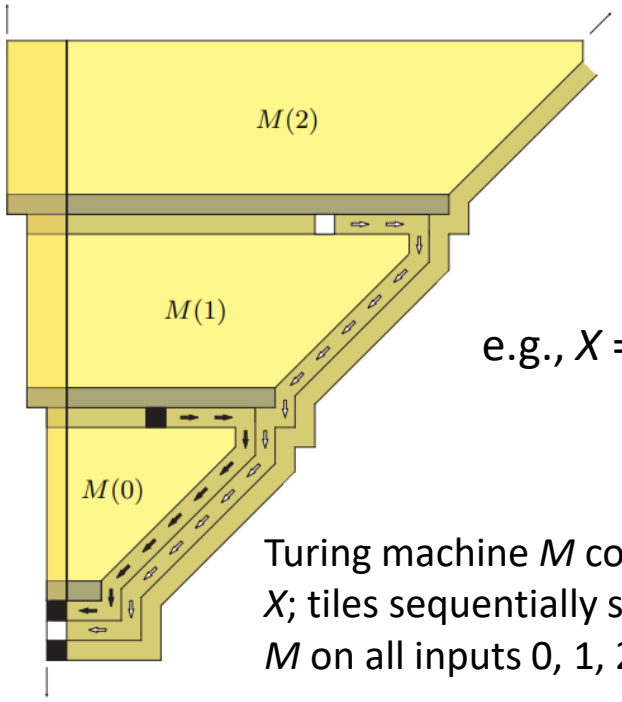

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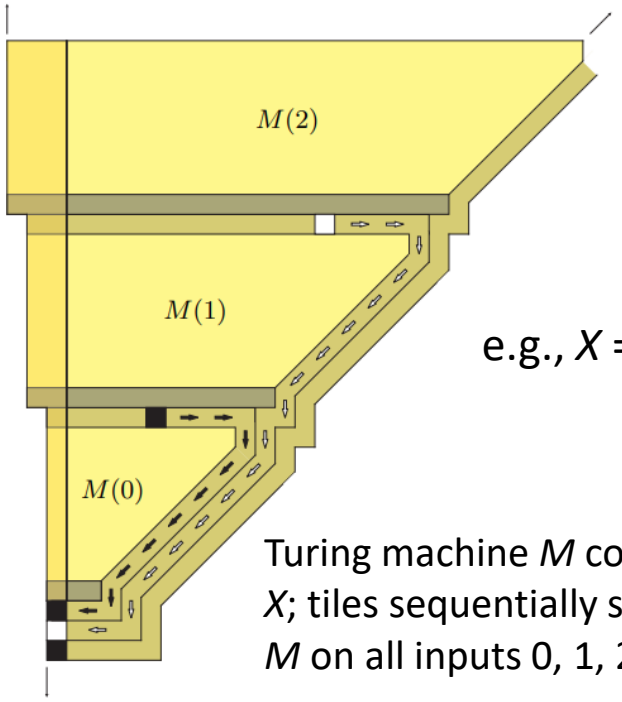

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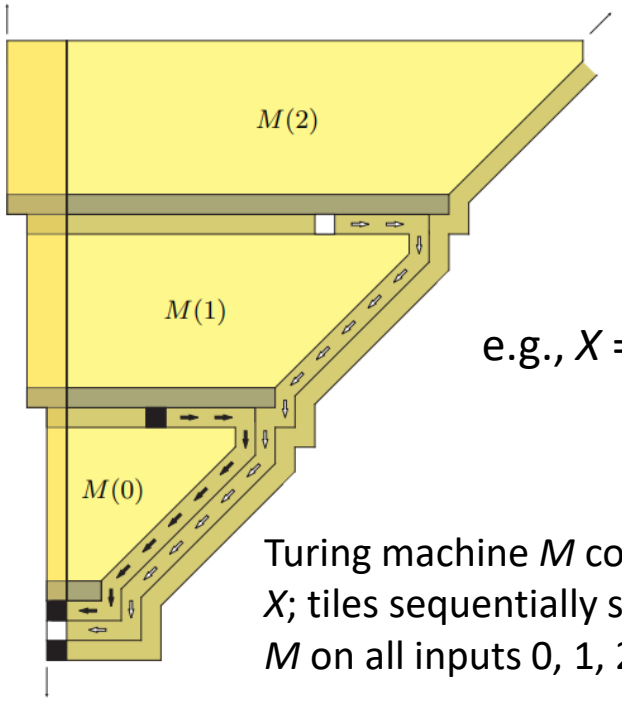

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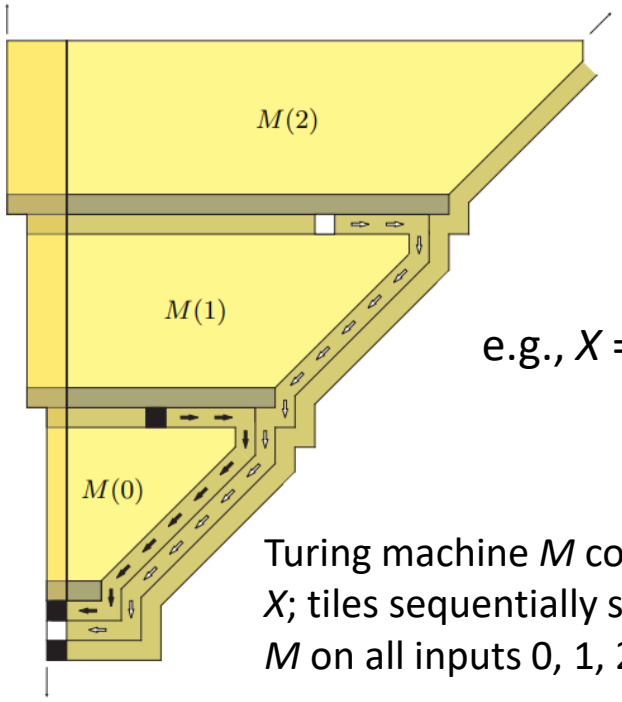

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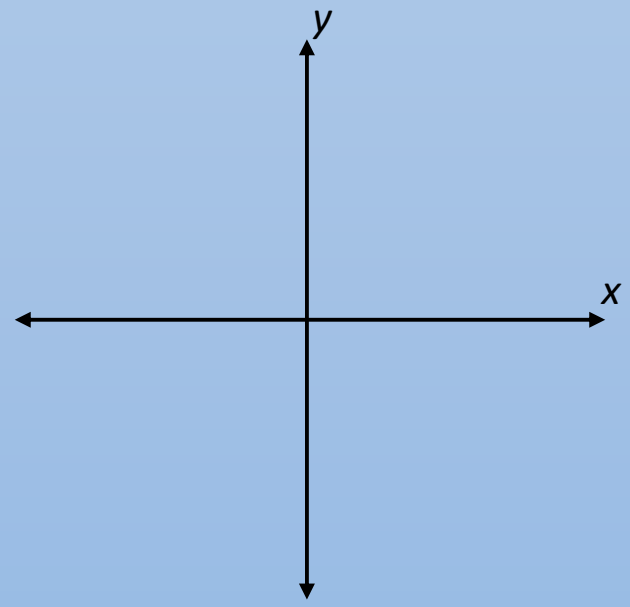

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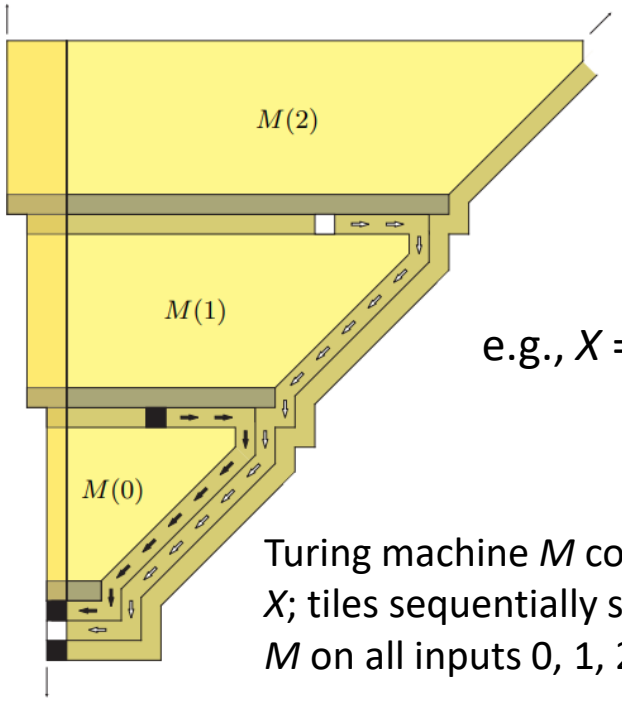

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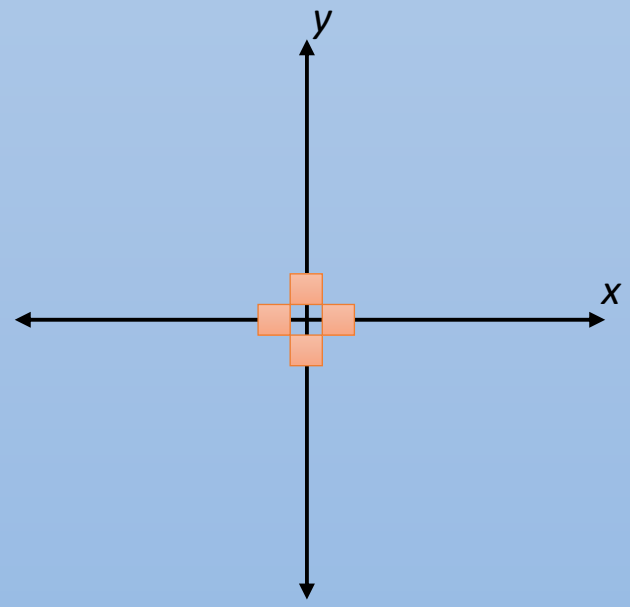

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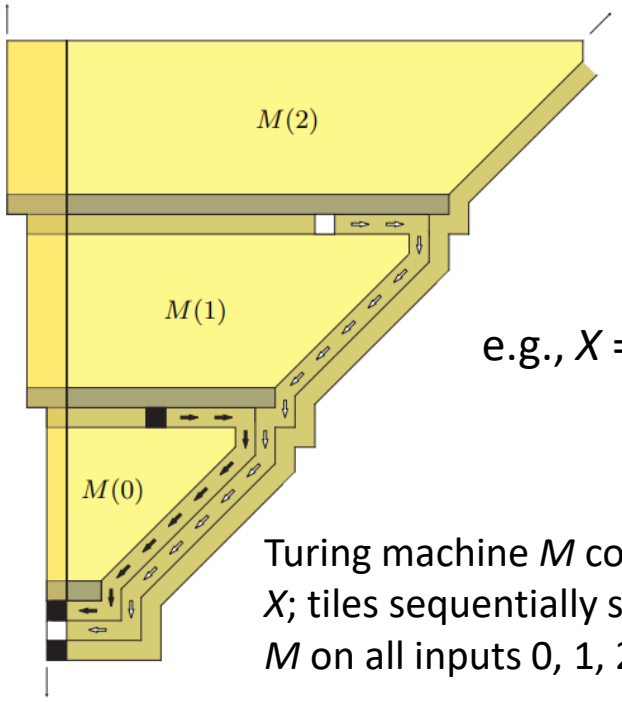

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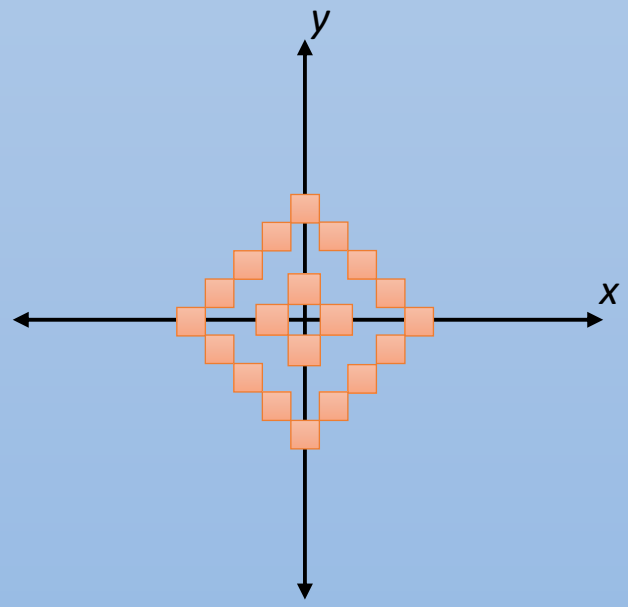

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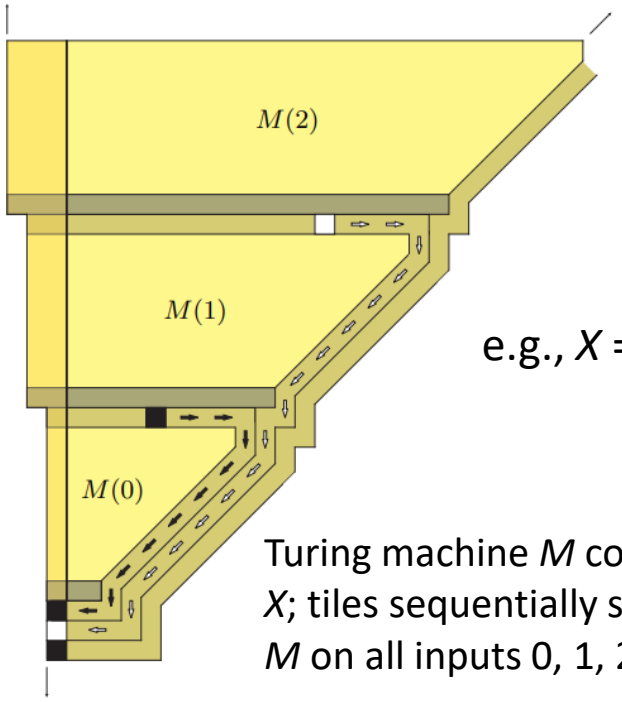

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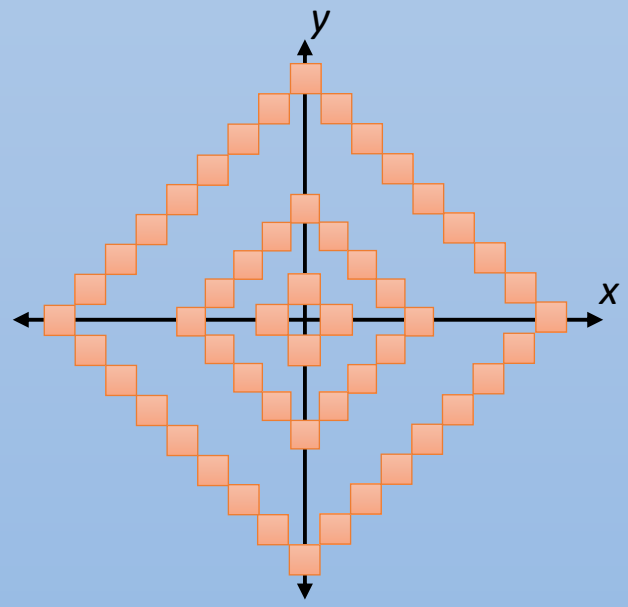

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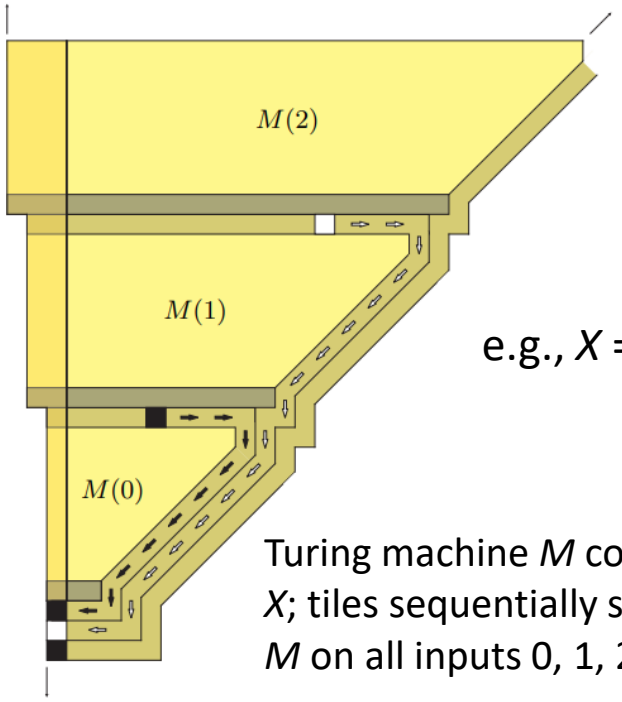

e.g., $X = \{0, 2, \ldots\}$

Turing machine M computes X; tiles sequentially simulate M on all inputs 0, 1, 2, …,

[Patitz, Summers, *Self-assembly of decidable sets*, UCNC 2008.]

Proof:

1. The Time Hierarchy Theorem says there is a computable set $A \subseteq \{1\}^*$ not computable in $O(n^4)$ time.
2. Let $R = \{|x| : x \in A\}$ be the set of lengths of strings in A.
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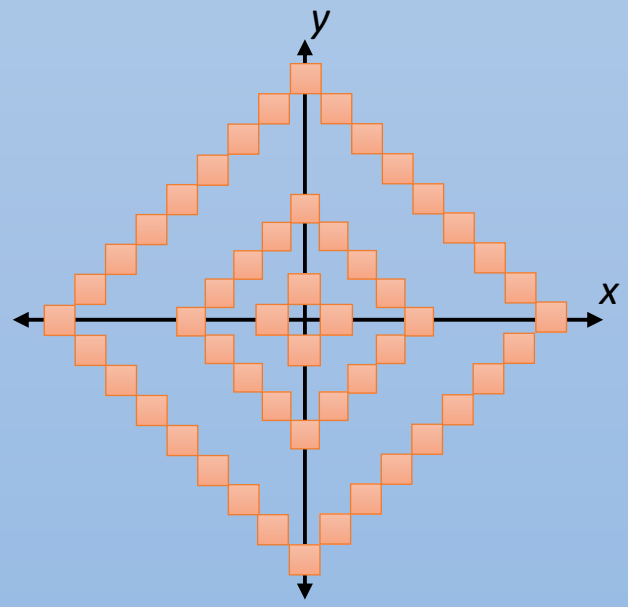

4. Suppose X could be weakly self-assembled. Then simulating self-assembly for $(2n)^2$ steps necessarily places a tile at some point at L_1 radius n from the origin; the tile's color tells us whether $n \in R \Leftrightarrow 1^n \in A$.

[Lathrop, Lutz, Patitz, Summers, *Computability and Complexity in Self-Assembly*, CiE 2008.]

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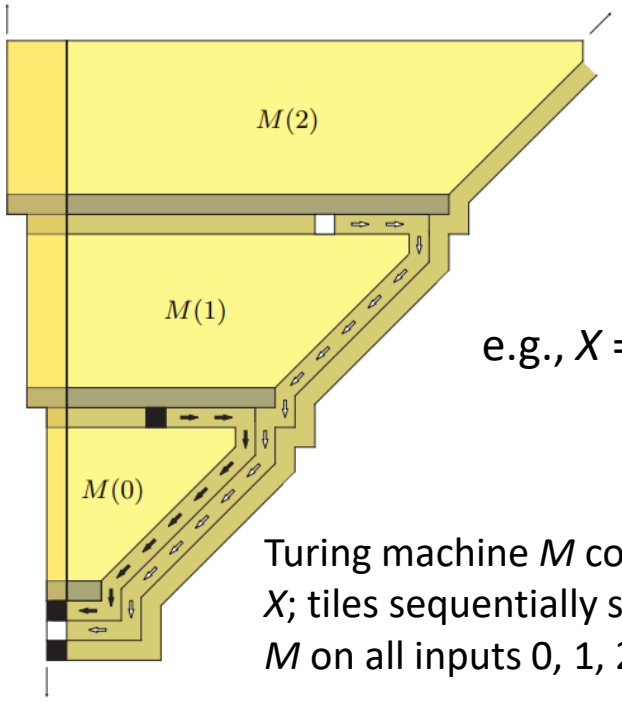

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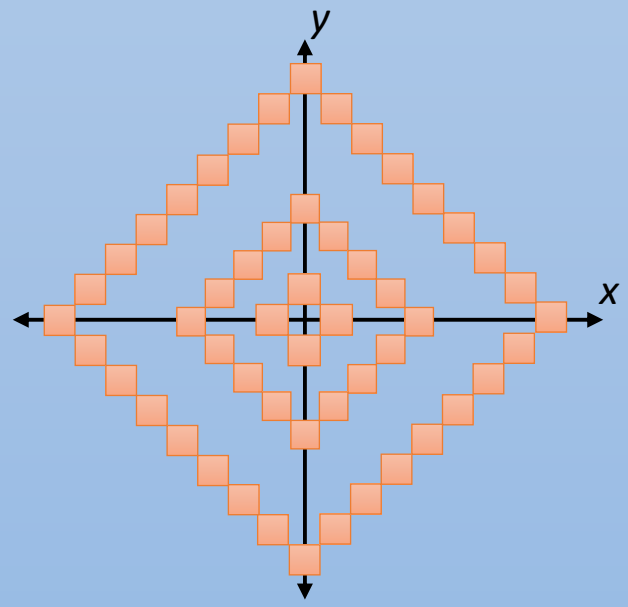

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5. This can be done in time $O(n^4)$ time (why?), a contradiction. **QED**

[Lathrop, Lutz, Patitz, Summers, *Computability and Complexity in Self-Assembly*, CiE 2008.]

Randomized self-assembly

Tile complexity of universal shape construction

- Recall: if we can have a seed structure encoding a shape S (in a binary string $x \in \{0,1\}^*$, in glues on one side), we can self-assemble some scaling S^c of S with $O(1)$ additional tile types that read and interpret x.

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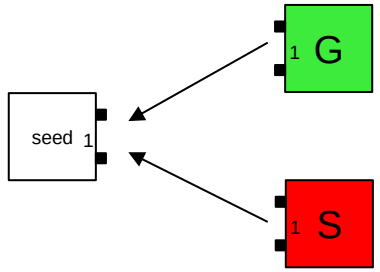
  - i.e., move the effort from designing new tile types to (*the plausibly simpler lab step of*) altering concentrations of existing tile types

Nondeterministic binding



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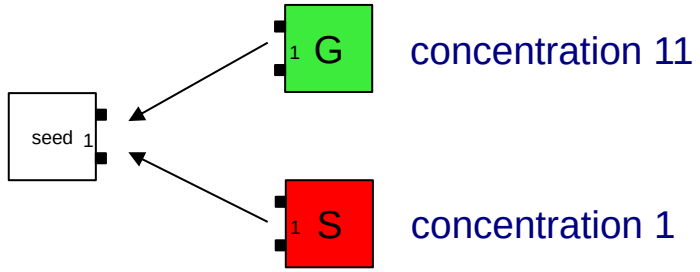

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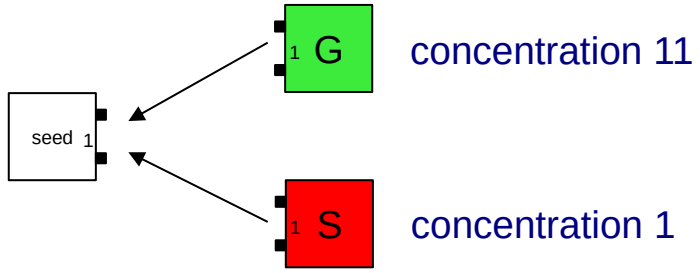
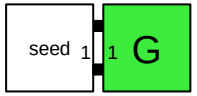
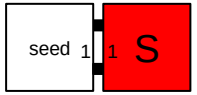


Pr[

] = 11/12

Pr[

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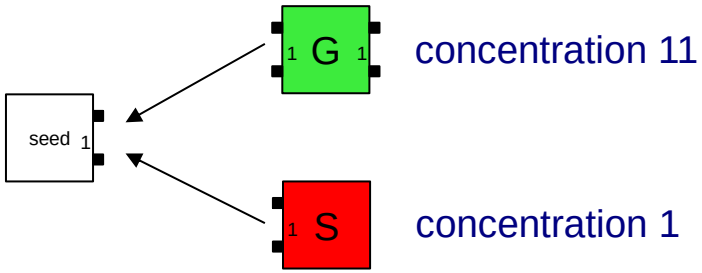
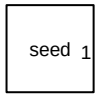
Programming polymer length with concentrations

[Becker, Rapaport, Rémila, *FSTTCS* 2006]





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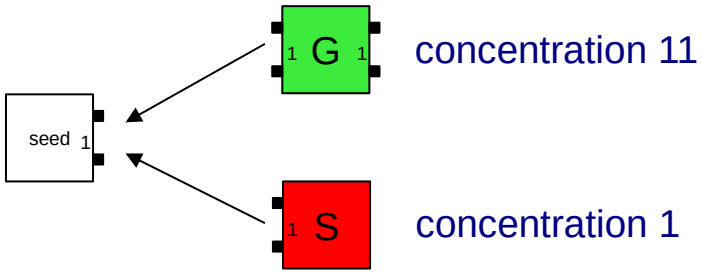

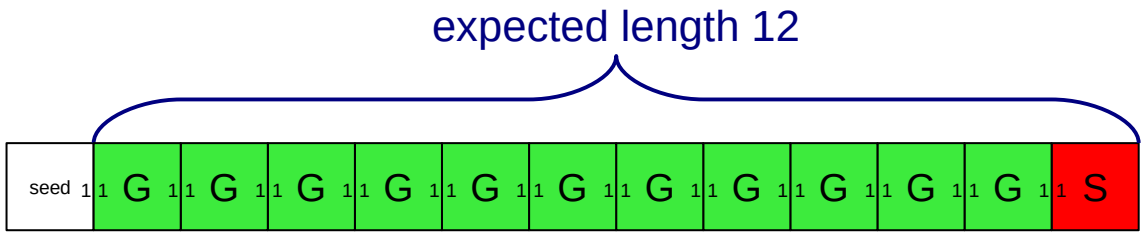

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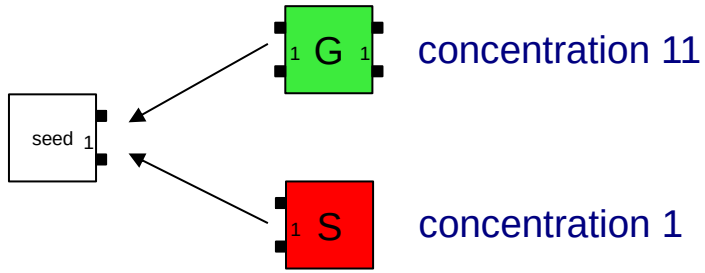
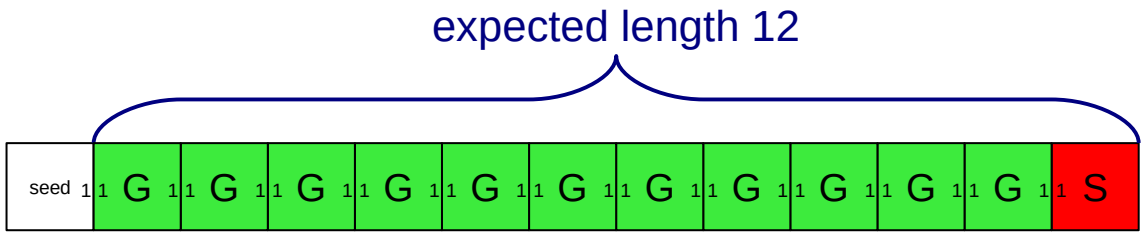
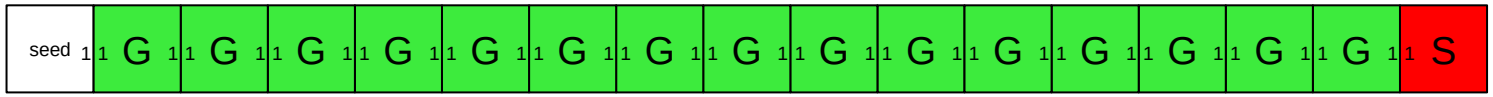
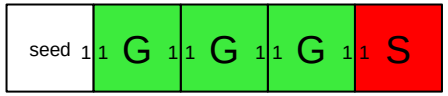


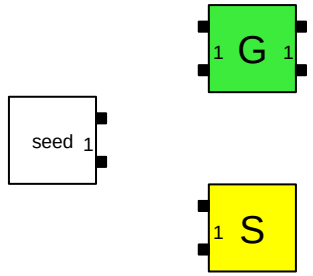






Large variance

Programming polymer length (improved)



concentration 3

concentration 1

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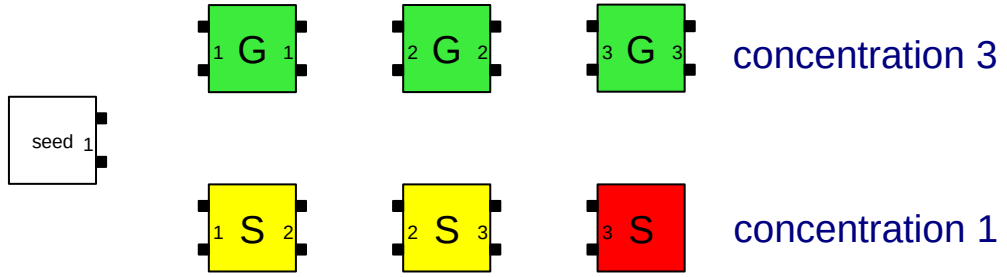
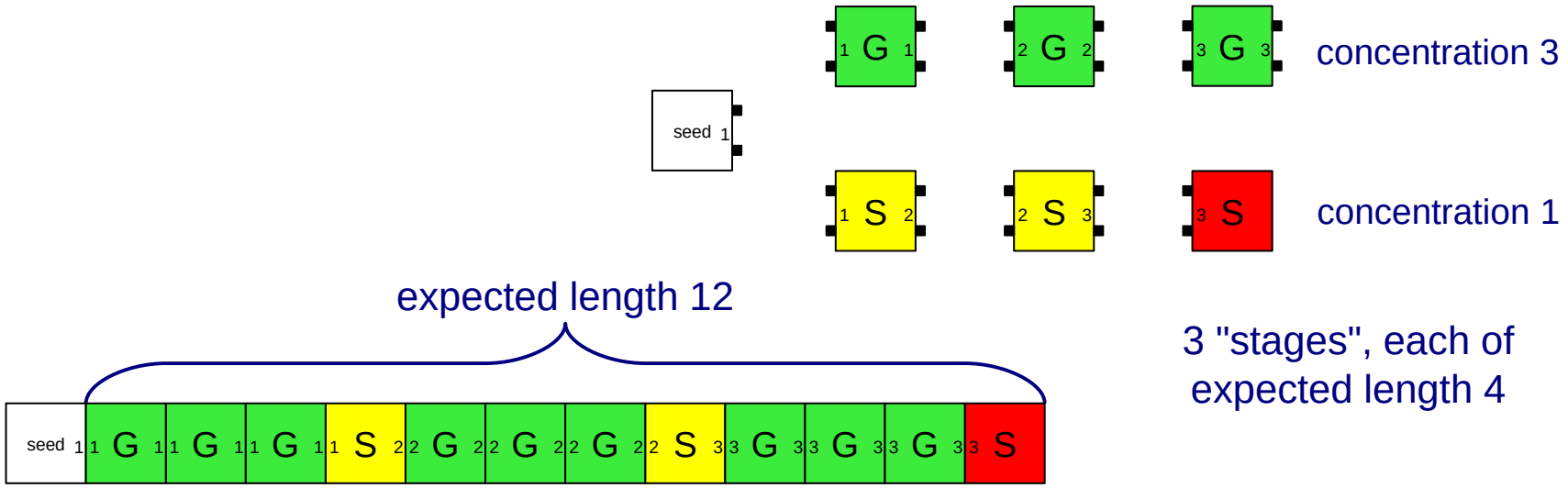


3 "stages", each of expected length 4

Programming polymer length (improved)



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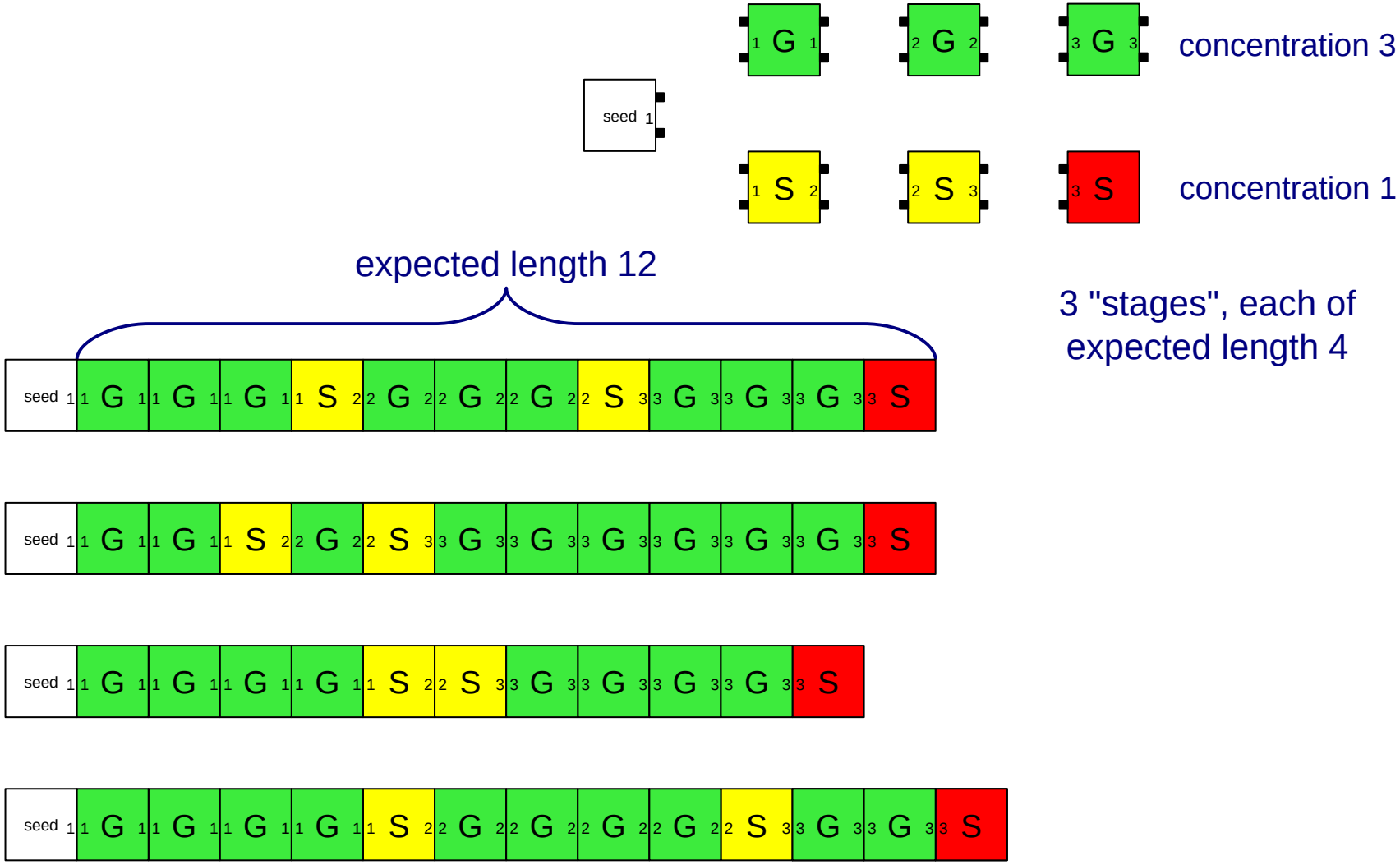

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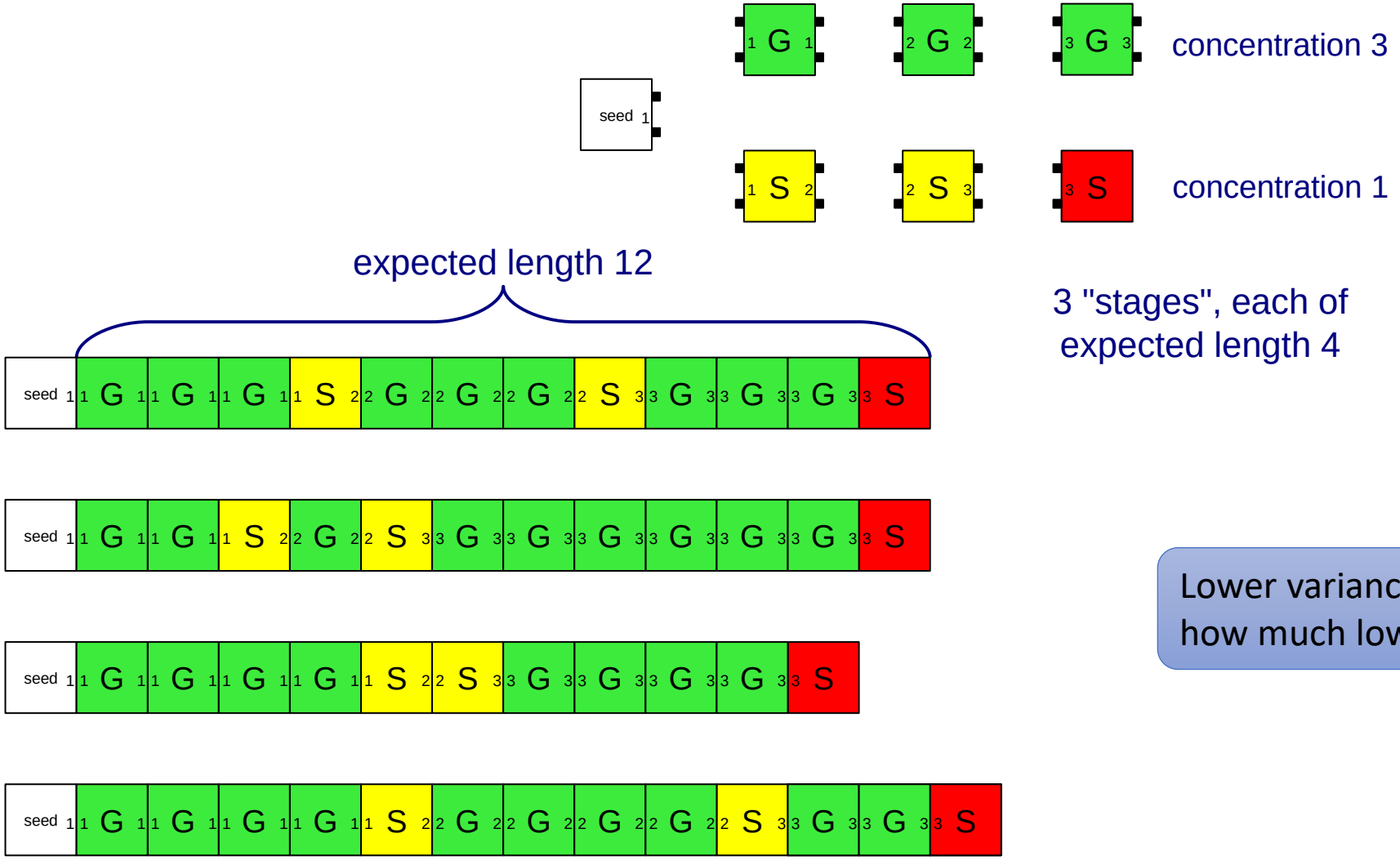

Bounding the probability the length deviates much from its mean

- *r* total stages, each with Pr[next tile 

increments stage] = *p*.

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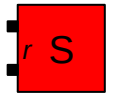
- Let **L**(*r*,*p*) = total length; number of tile attachments until attaching 

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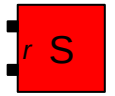
- Let **L**(*r*,*p*) = total length; number of tile attachments until attaching 

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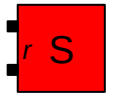
- Let **L**(*r*,*p*) = total length; number of tile attachments until attaching 
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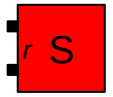
- Let $\mathbf{L}(r,p)$ = total length; number of tile attachments until attaching 

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- for any n,r,p: $\Pr[\mathbf{L}(r,p) \leq n] = \Pr[\mathbf{B}(n,p) \geq r]$

flipping a coin until the r'th heads requires $\leq n$ flips $\Leftrightarrow$ flipping a coin n times results in $\geq r$ heads

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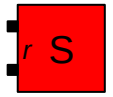
- Let $\mathbf{L}(r,p)$ = total length; number of tile attachments until attaching 
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Chernoff bound

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$\Pr[\mathbf{B}(n,p) > (1+\delta)np] < \exp(-\delta^2 np/3)$

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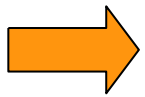
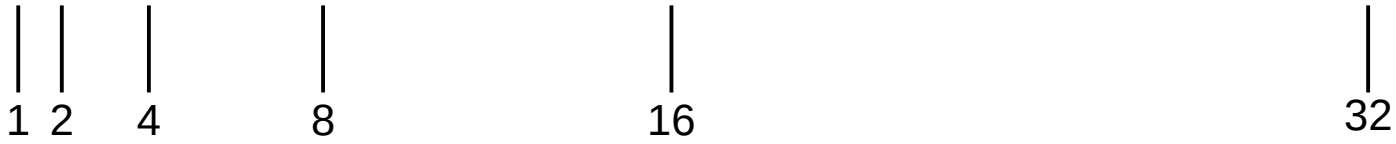
Let $\delta \approx 0.27$ and set p such that $r/p(1-\delta) = 2^k$.
Let $\delta' \approx 0.44$: then $r/p(1+\delta') \approx 2^{k-1}$.
Applying this to our setting gives
$\Pr[\mathbf{L}(r,p)$ is not between 2^{k-1} and $2^k] < 2 \cdot 0.9421^r$

Programming polymer length (improved)

if r = 90 stages, expected length midway in $[2^{k-1}, 2^k)$



with probability > 99%, **actual** length in $[2^{k-1}, 2^k)$



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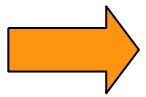

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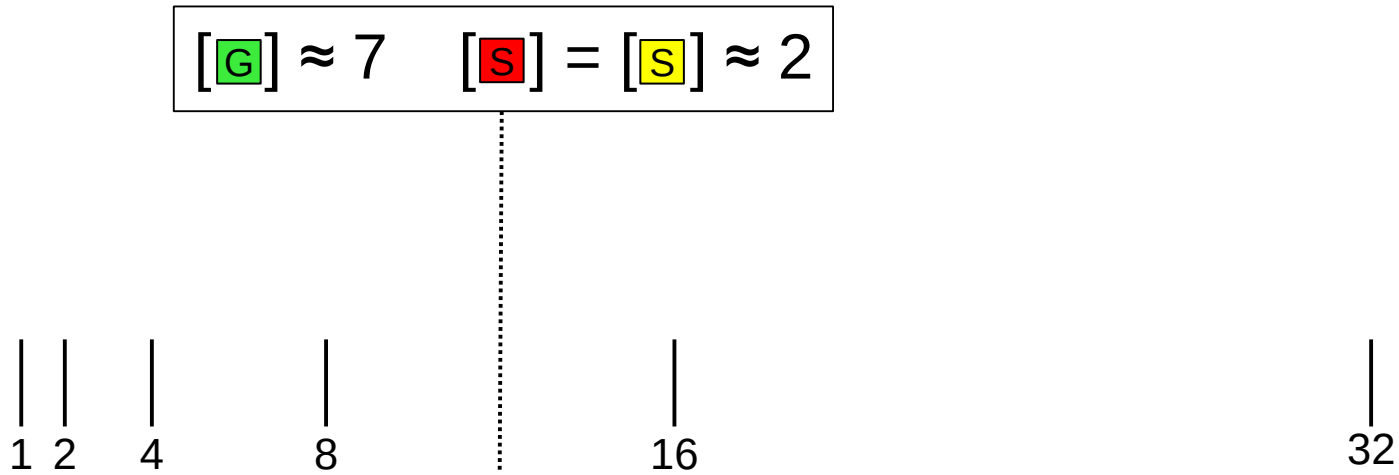

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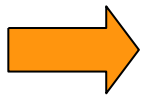

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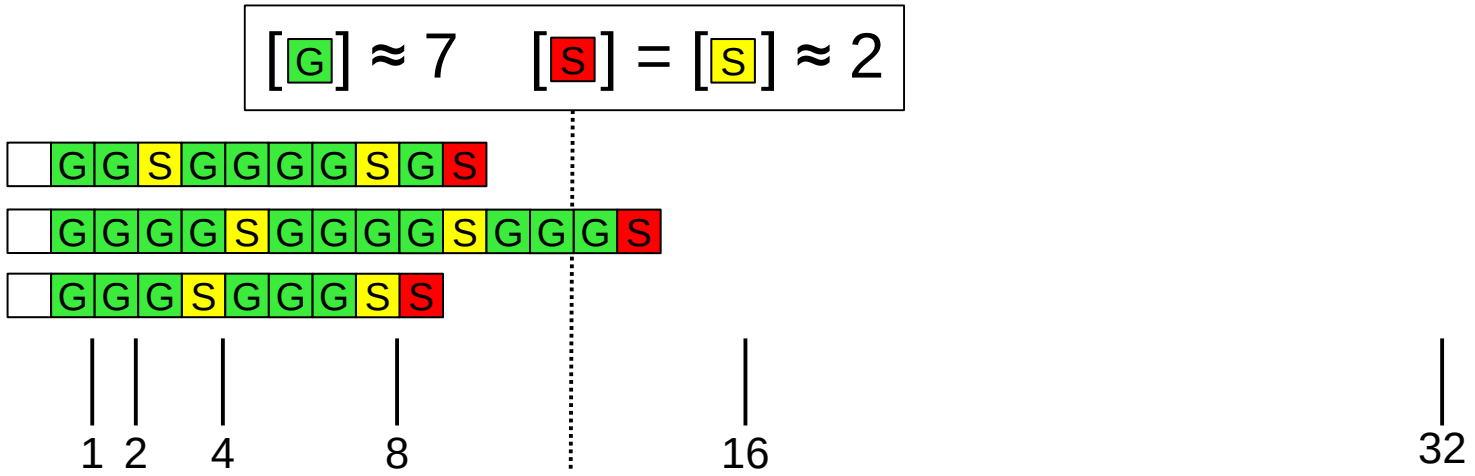

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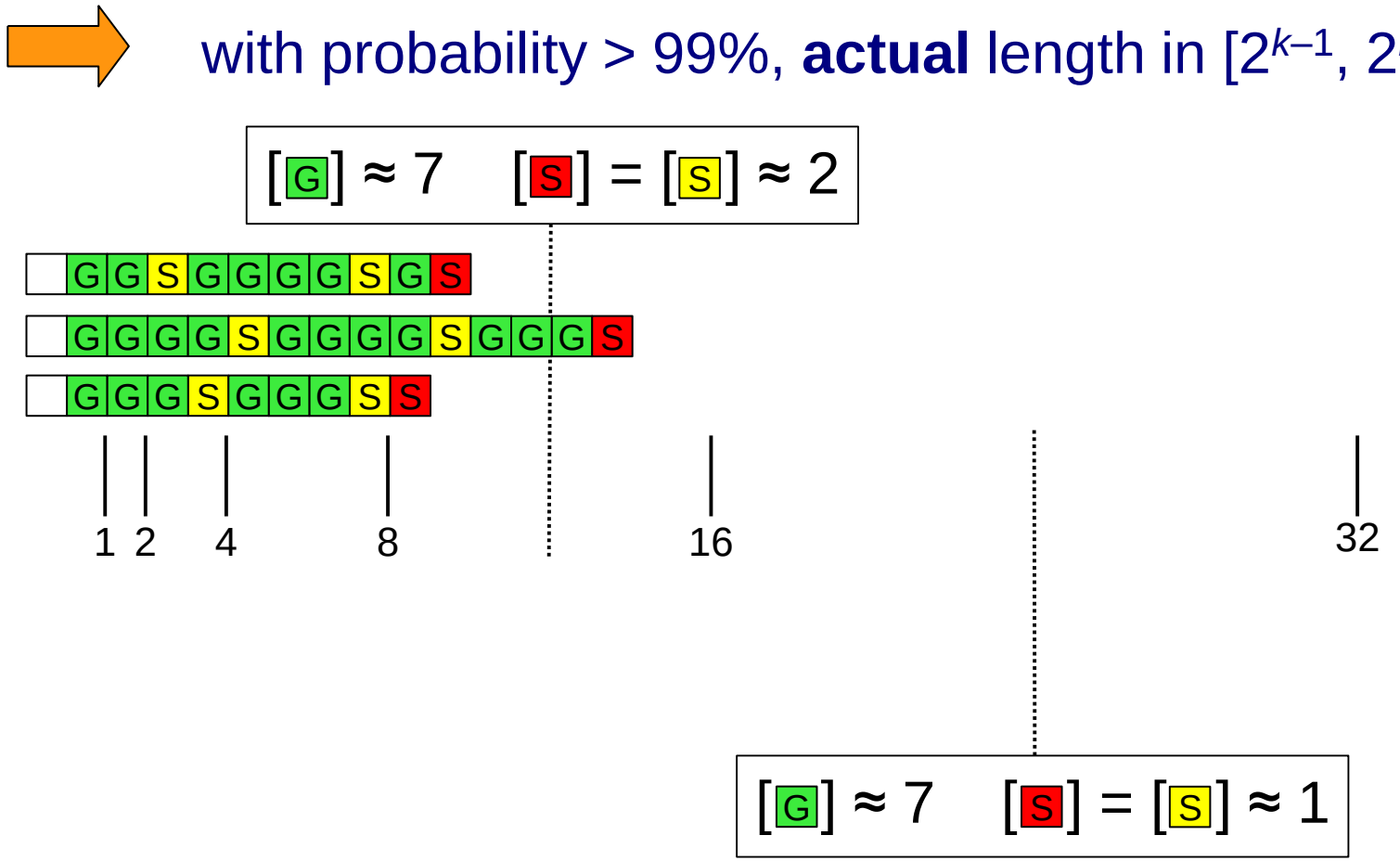

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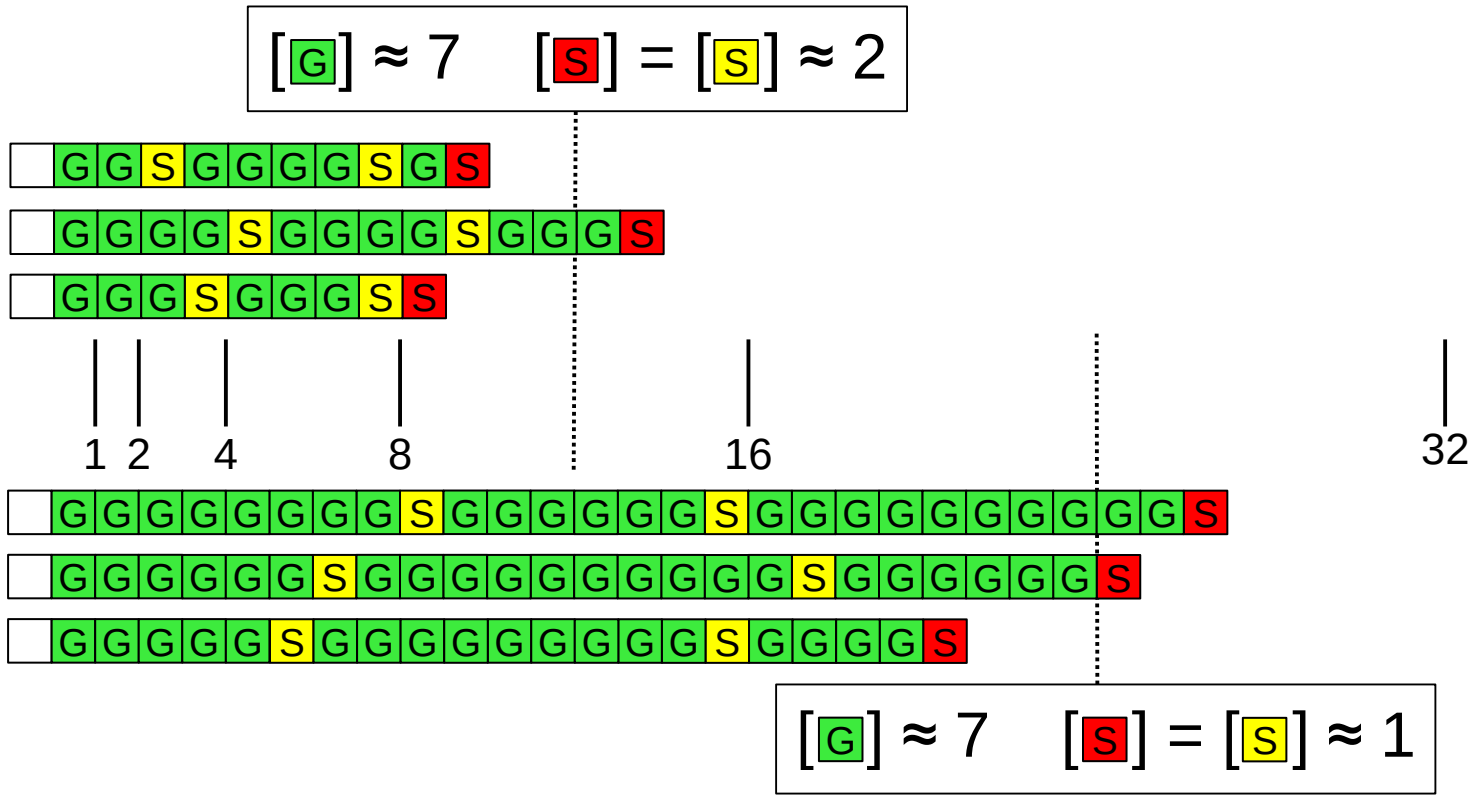
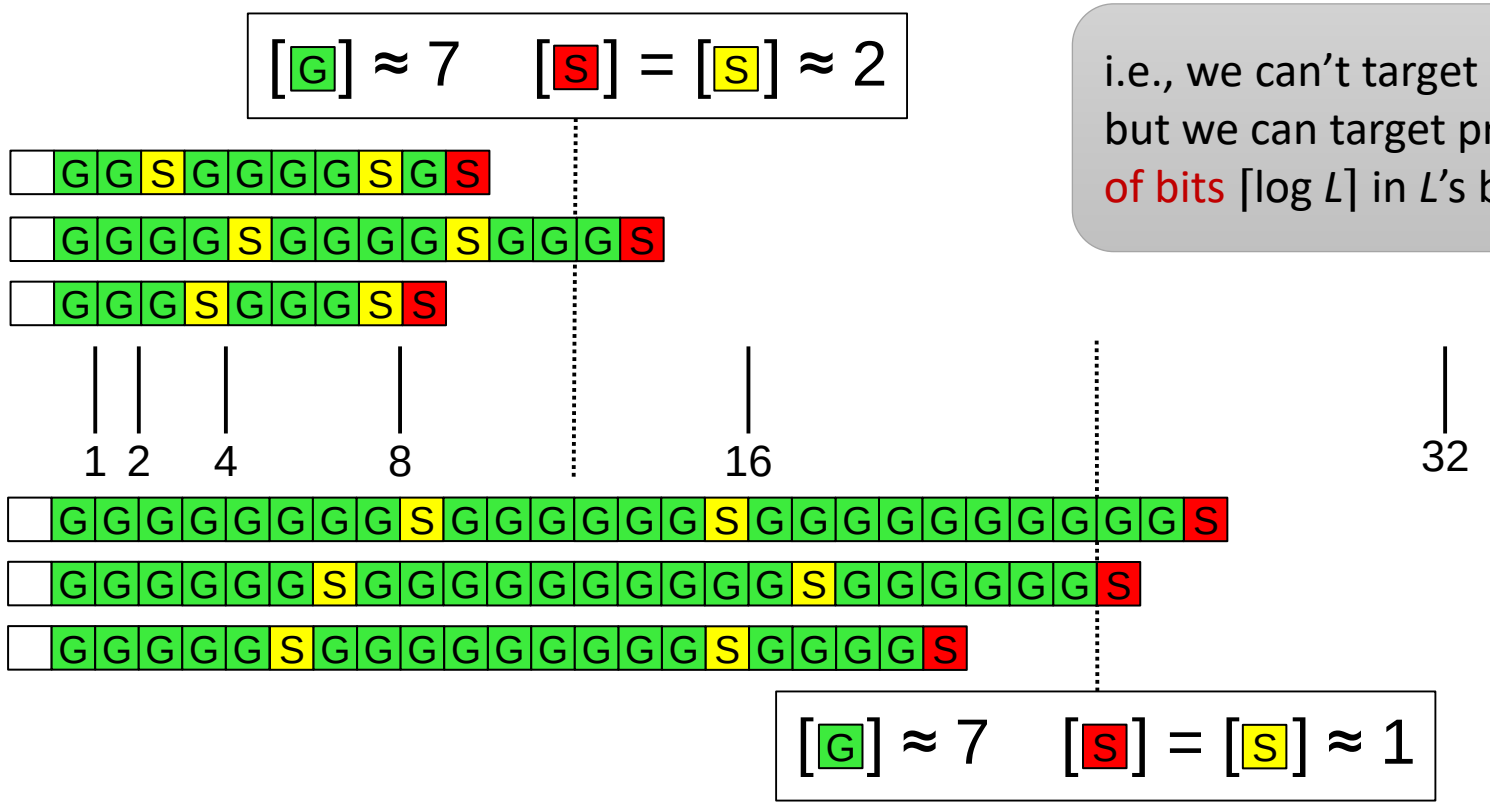


Programming polymer length (improved)

if r = 90 stages, expected length midway in $[2^{k-1}, 2^k)$

⇒ with probability > 99%, **actual** length in $[2^{k-1}, 2^k)$



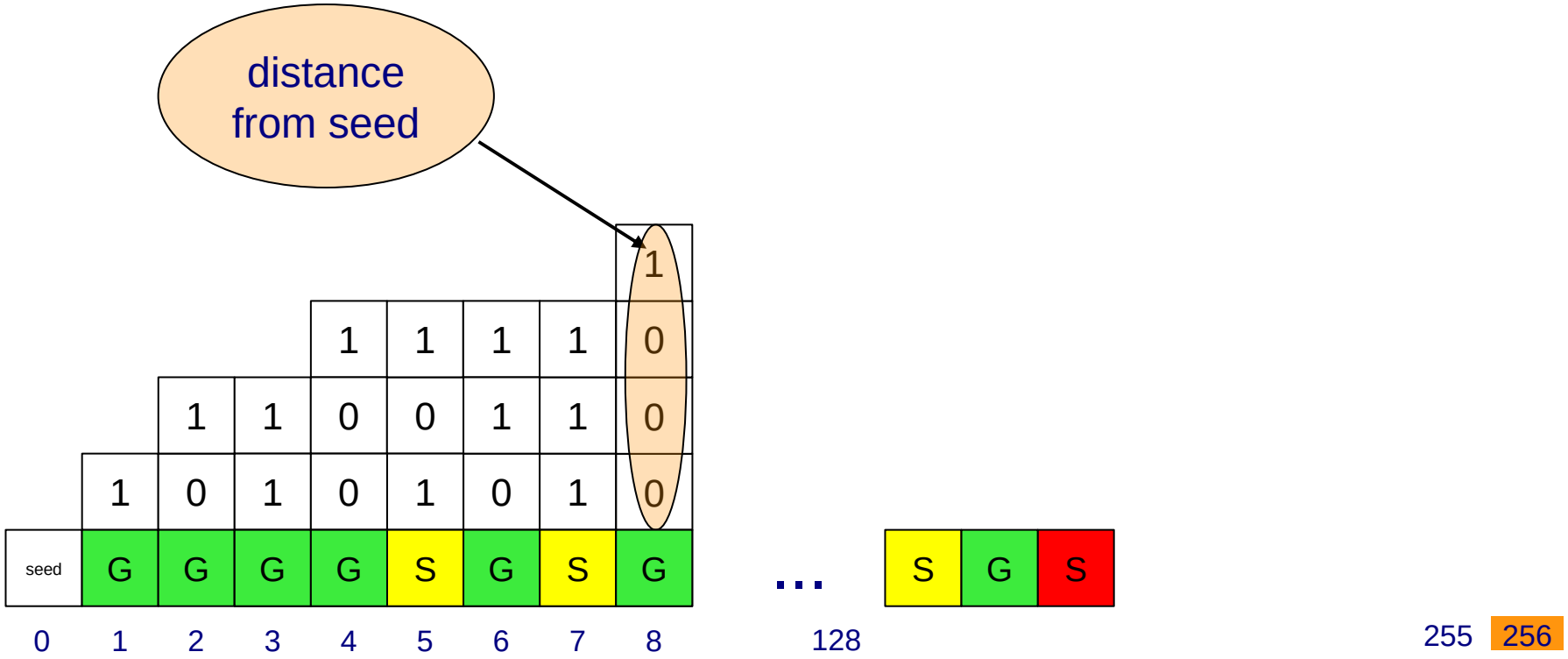
i.e., we can't target a precise length L, but we can target precisely the number of bits $\lceil \log L \rceil$ in L's binary expansion.

Programming polymer length 2^k precisely

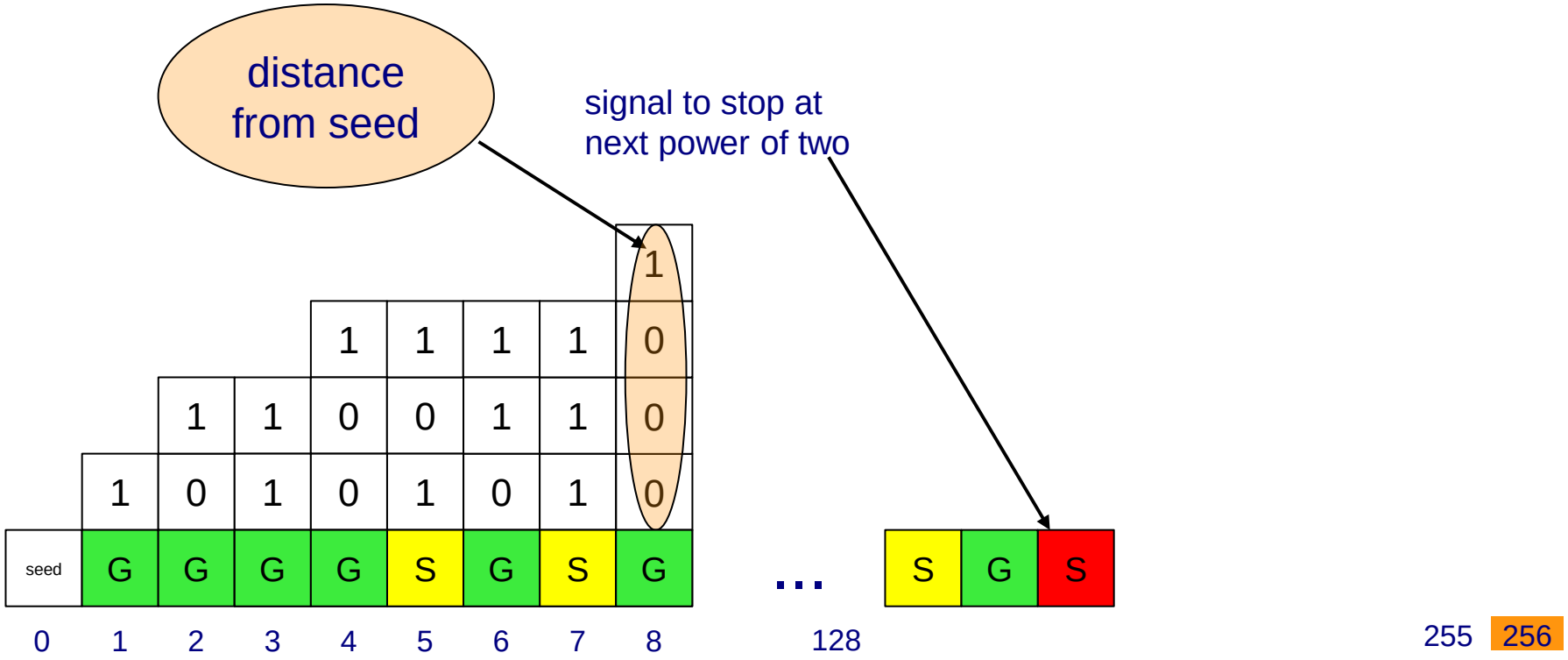
256

Programming polymer length 2^k precisely

Programming polymer length 2^k precisely



Programming polymer length 2^k precisely



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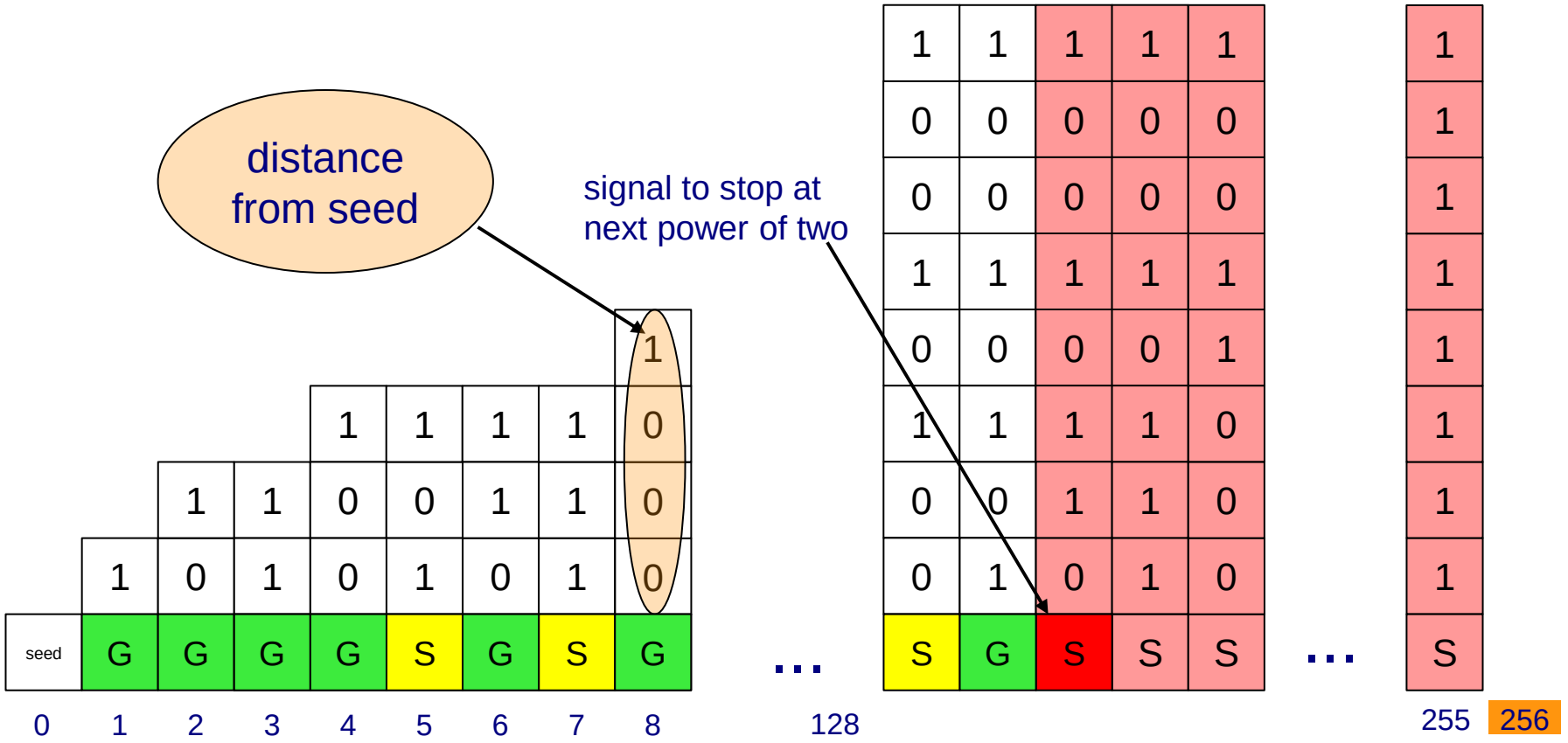

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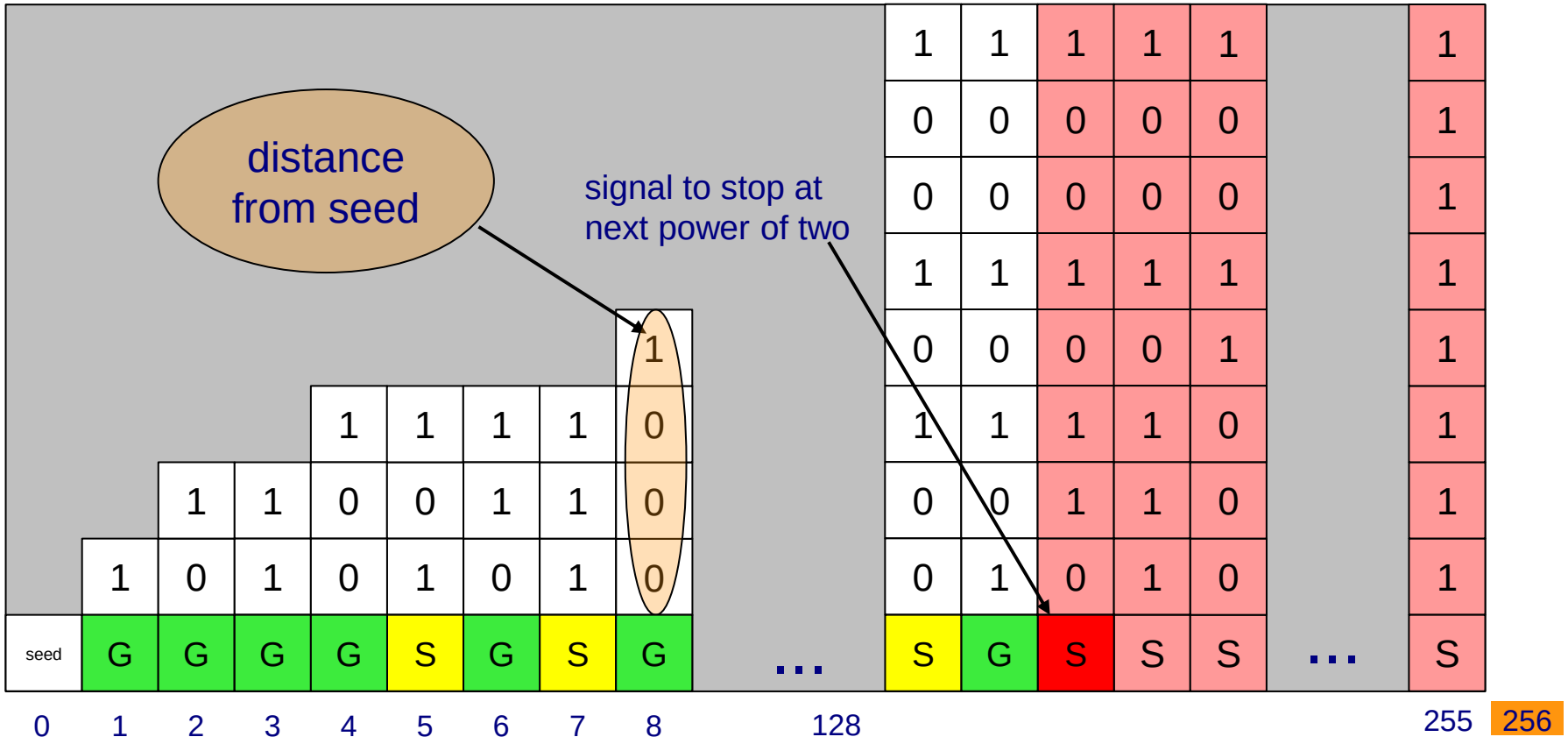
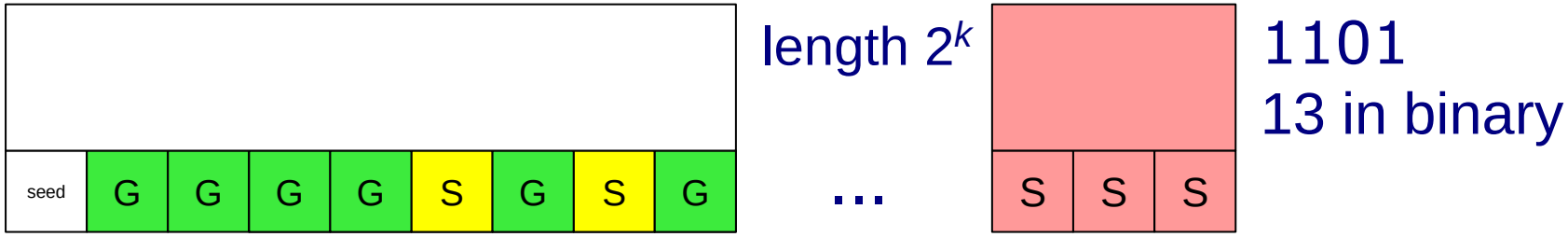


Programming a binary string

1101

13 in binary

Programming a binary string



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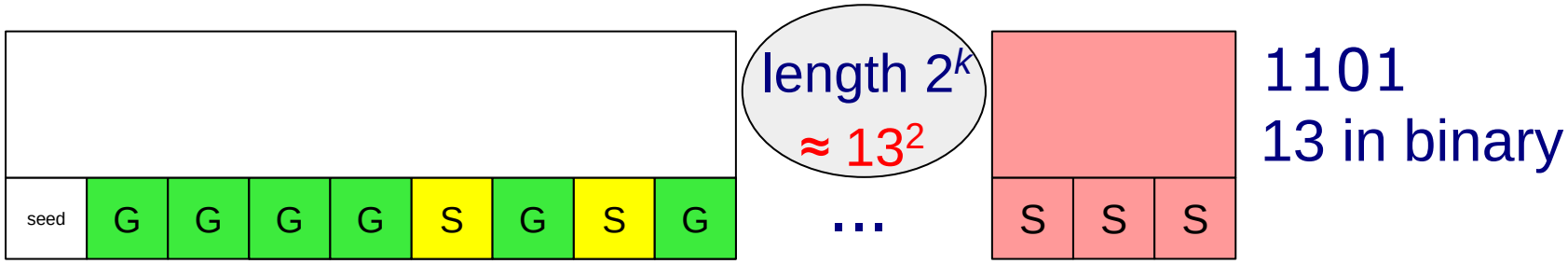

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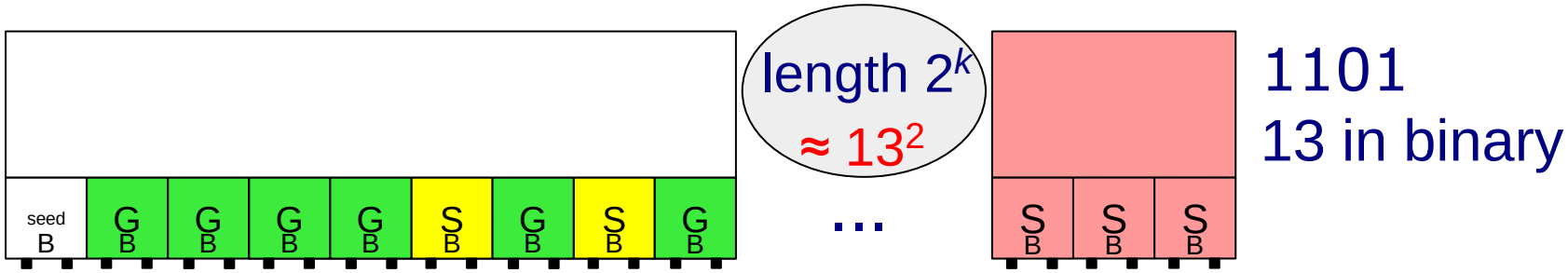
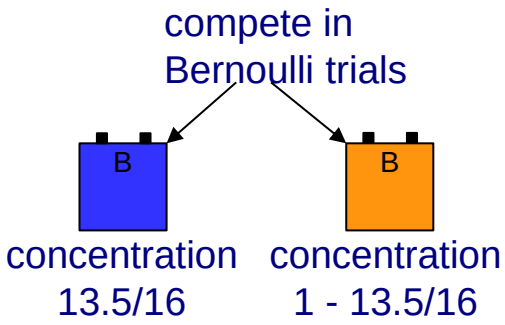
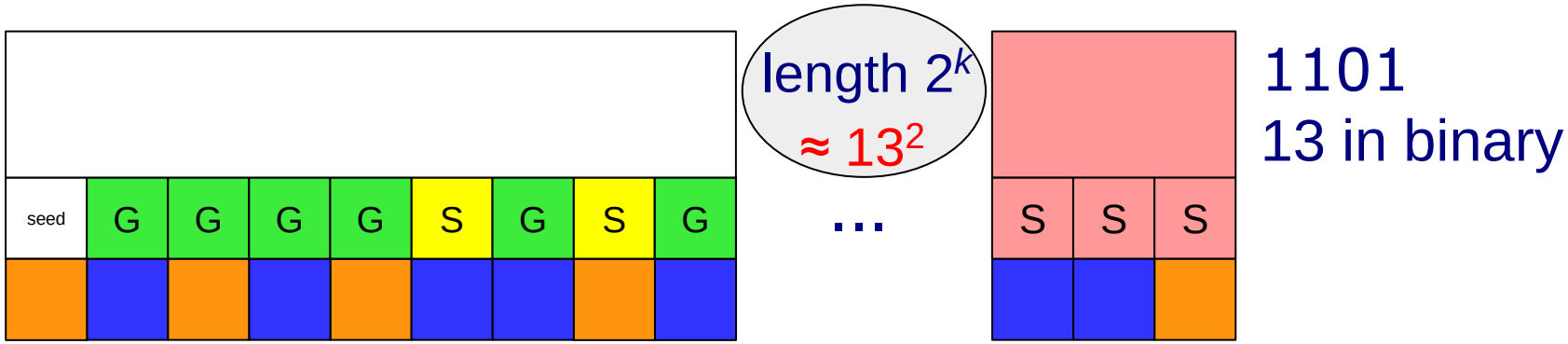
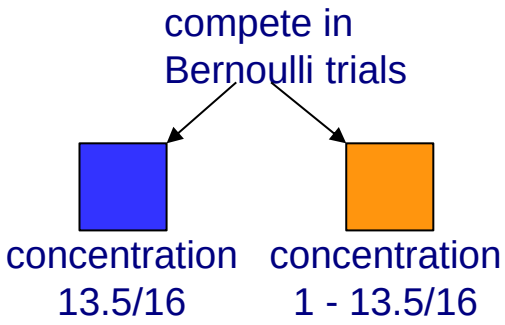




Programming a binary string





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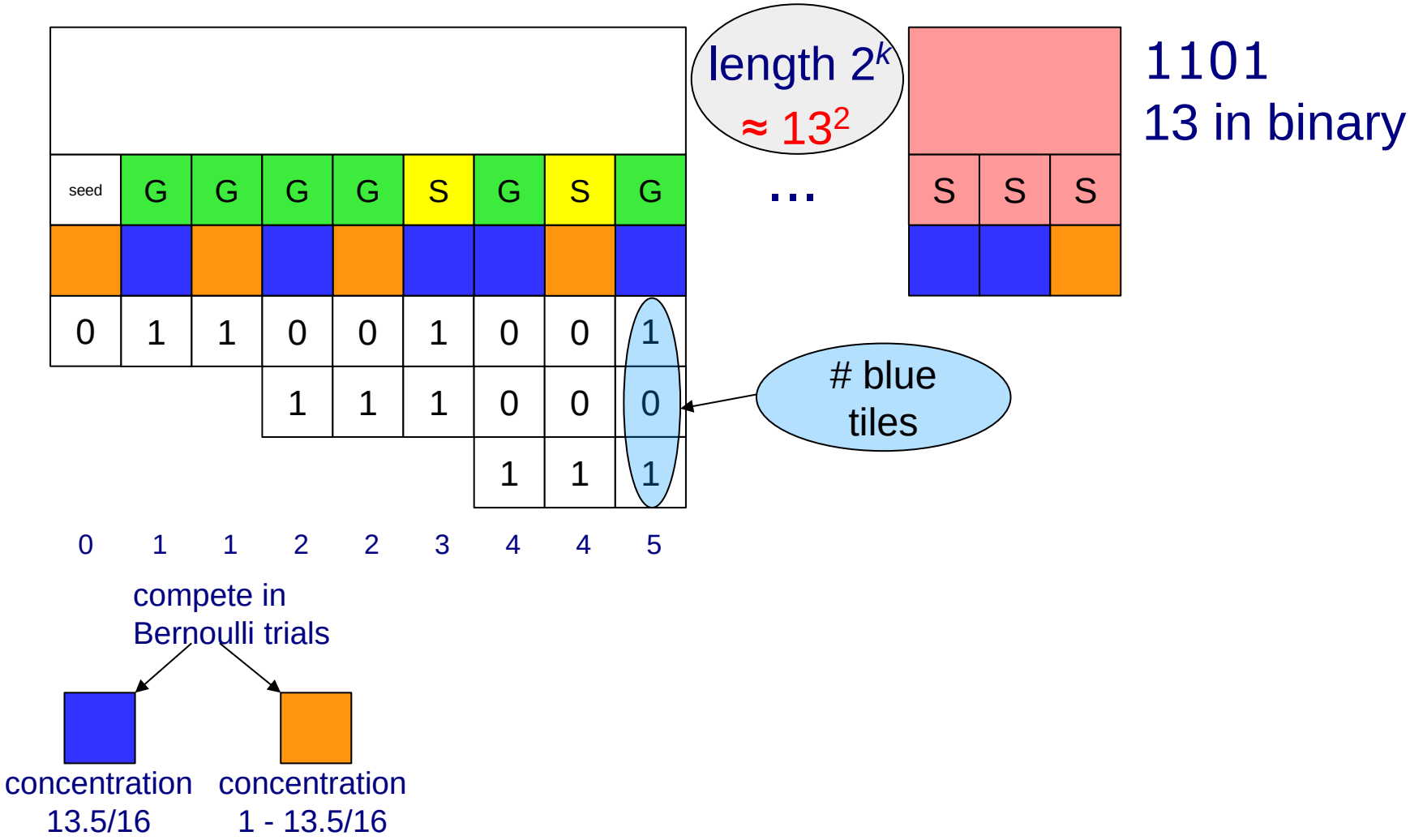

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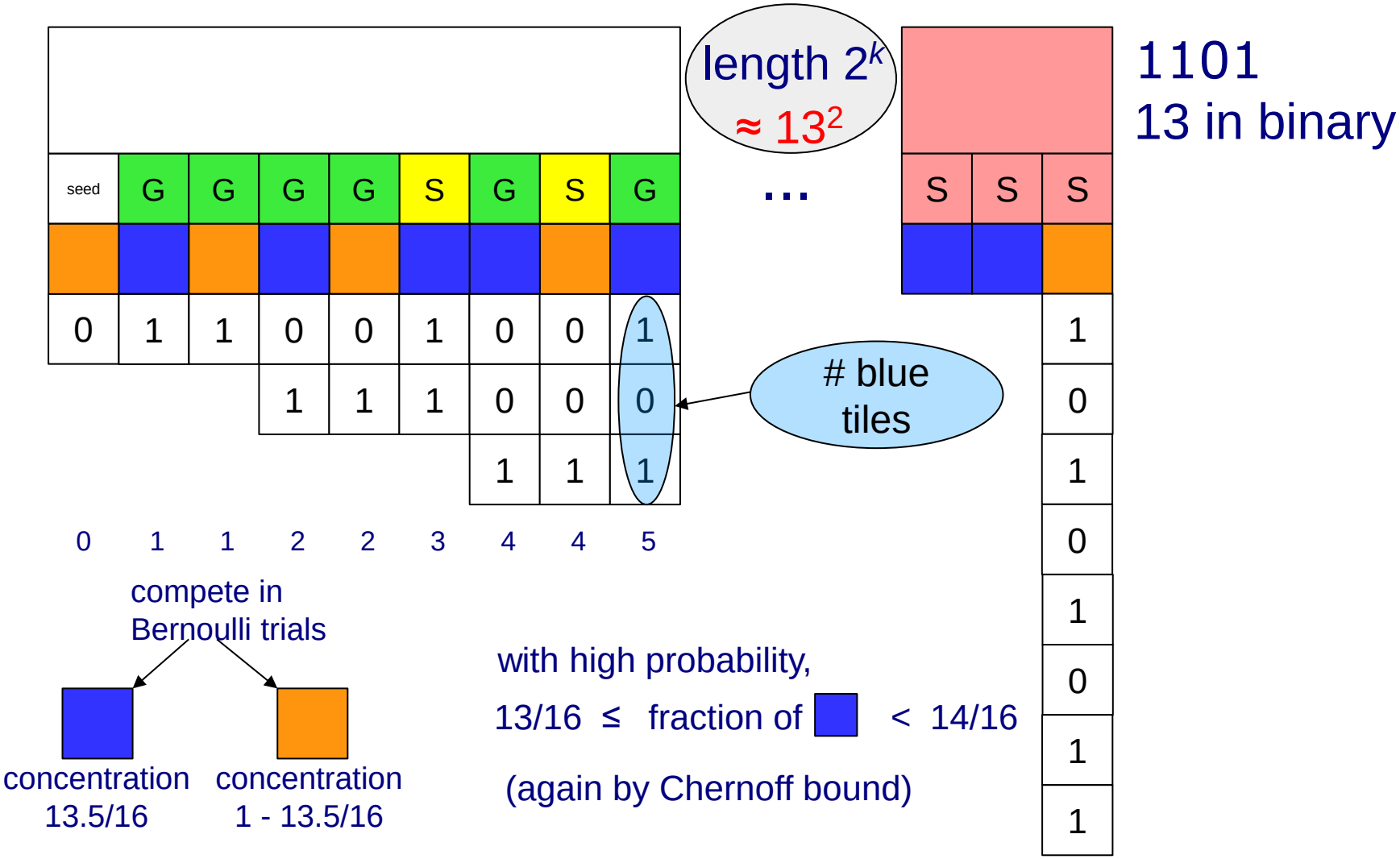

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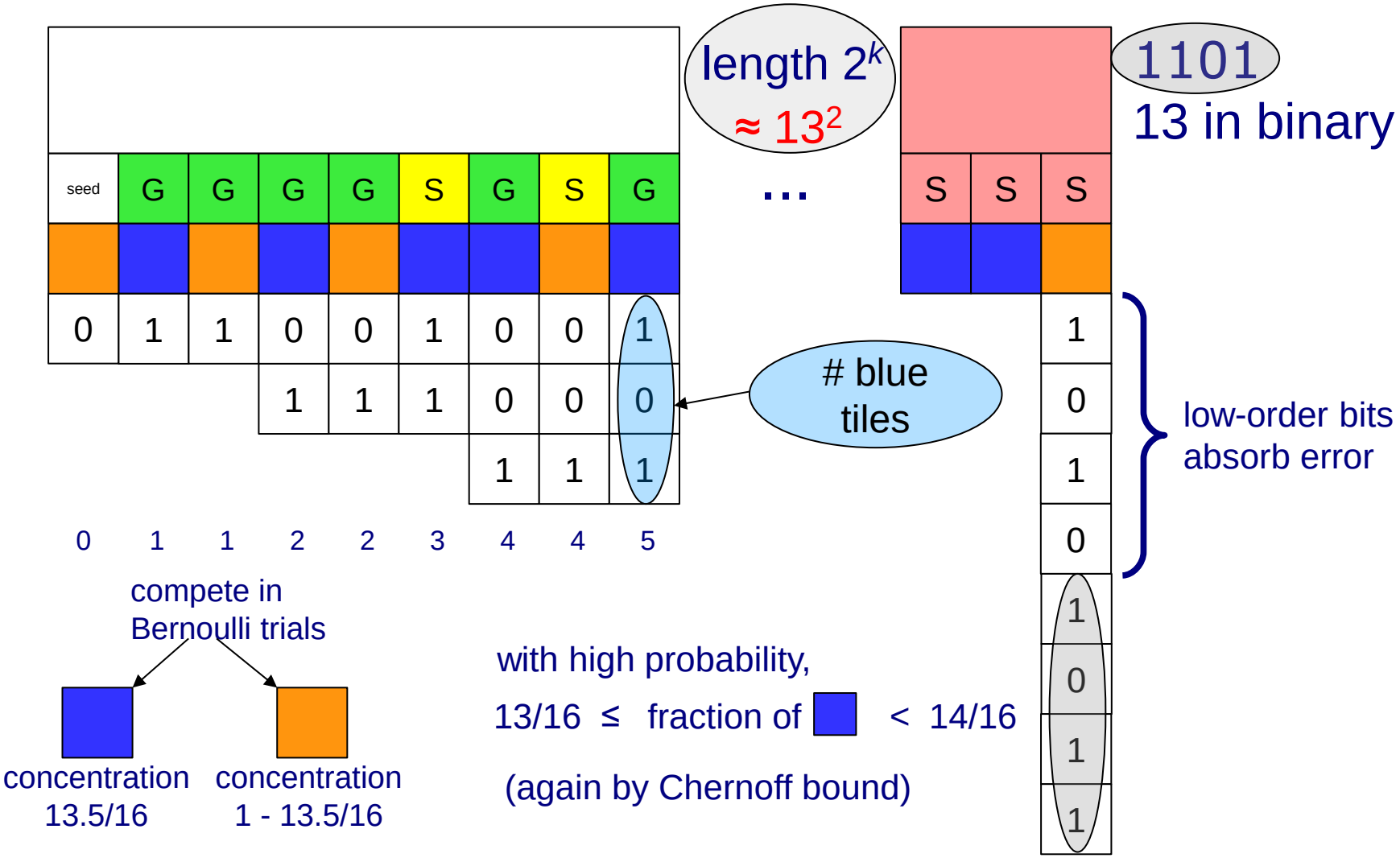

Programming a shape (inaccurate cartoonish overview)

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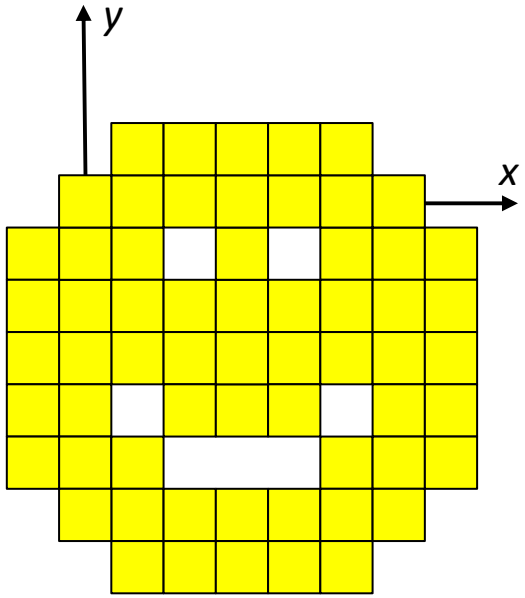

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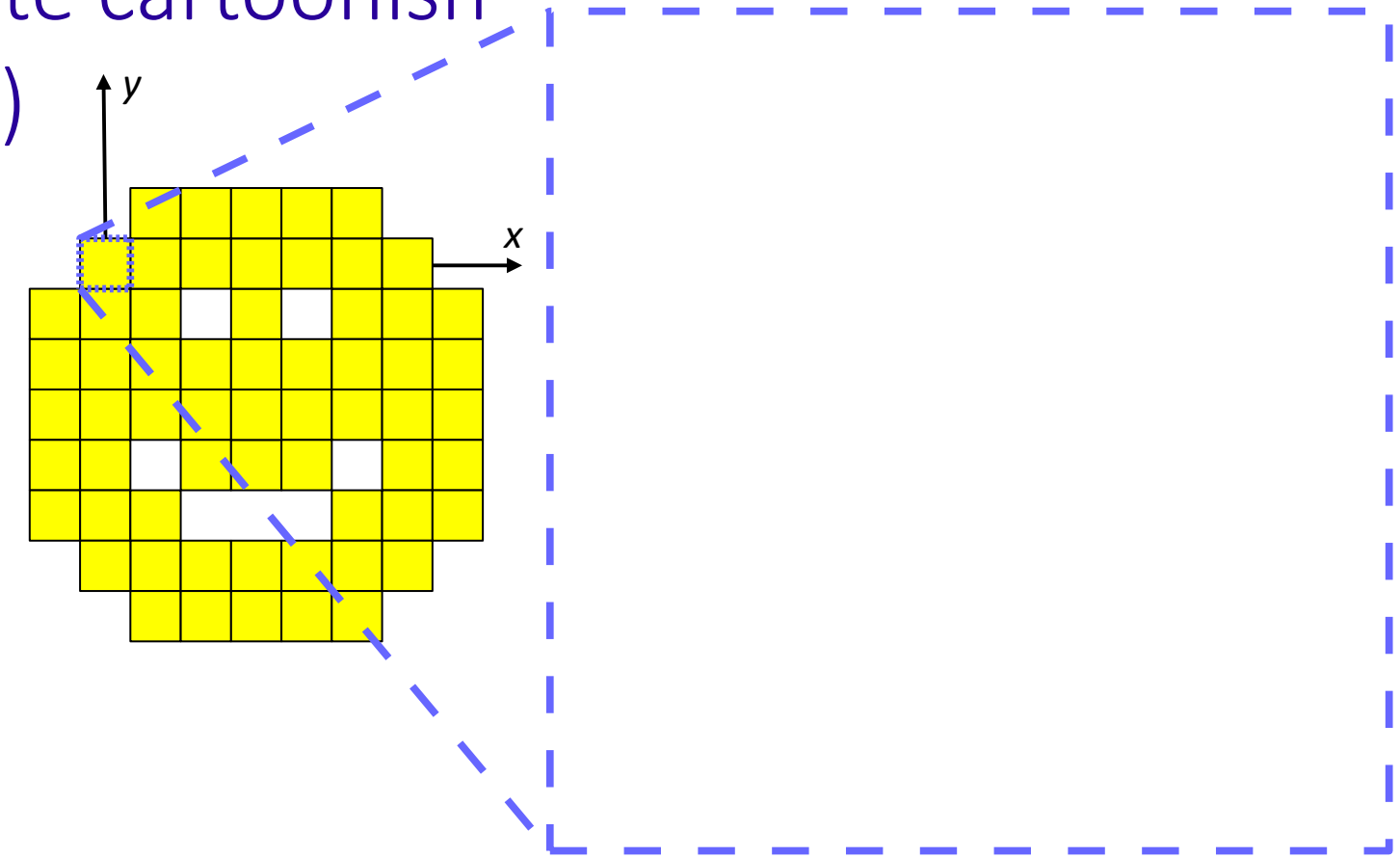

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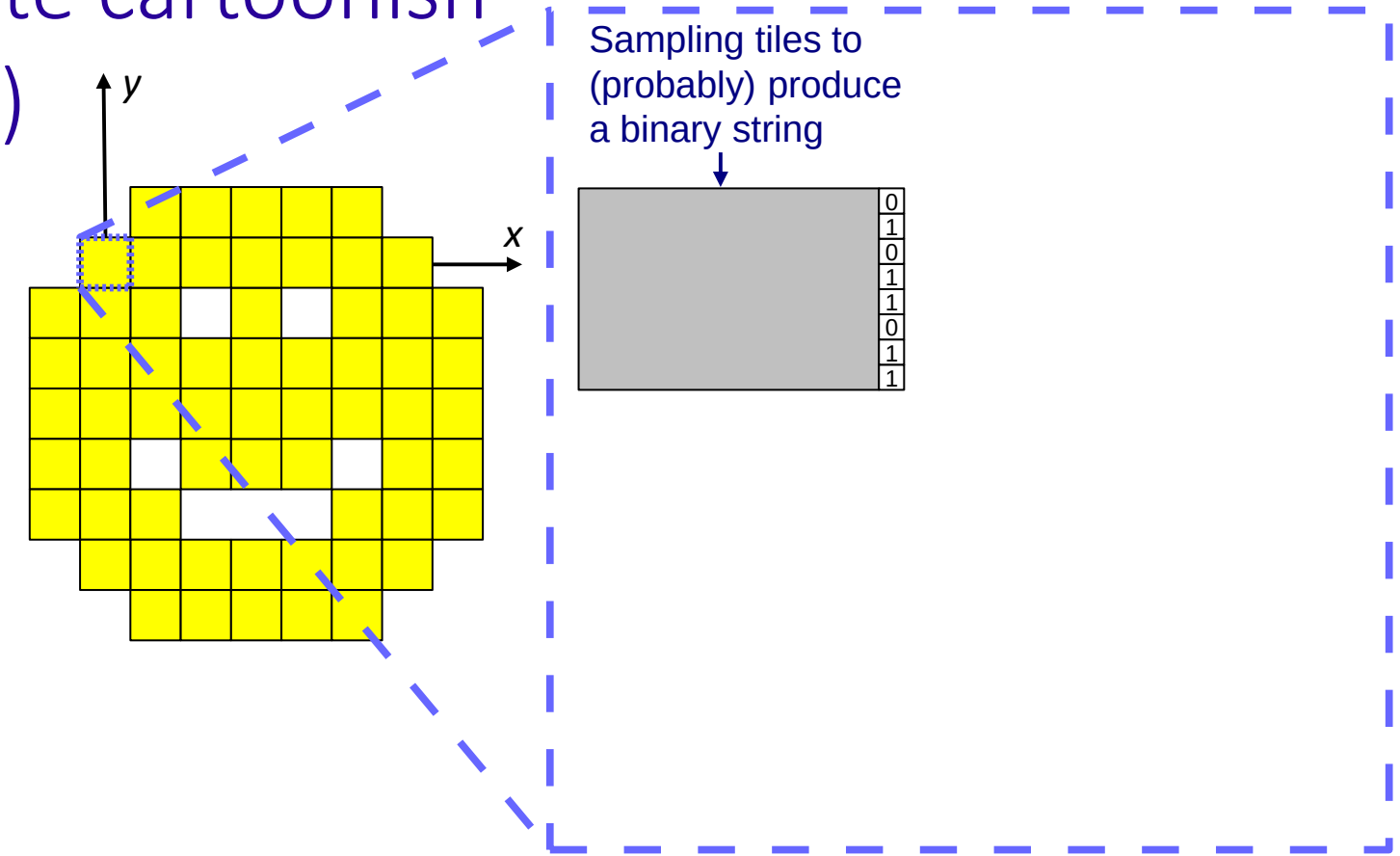

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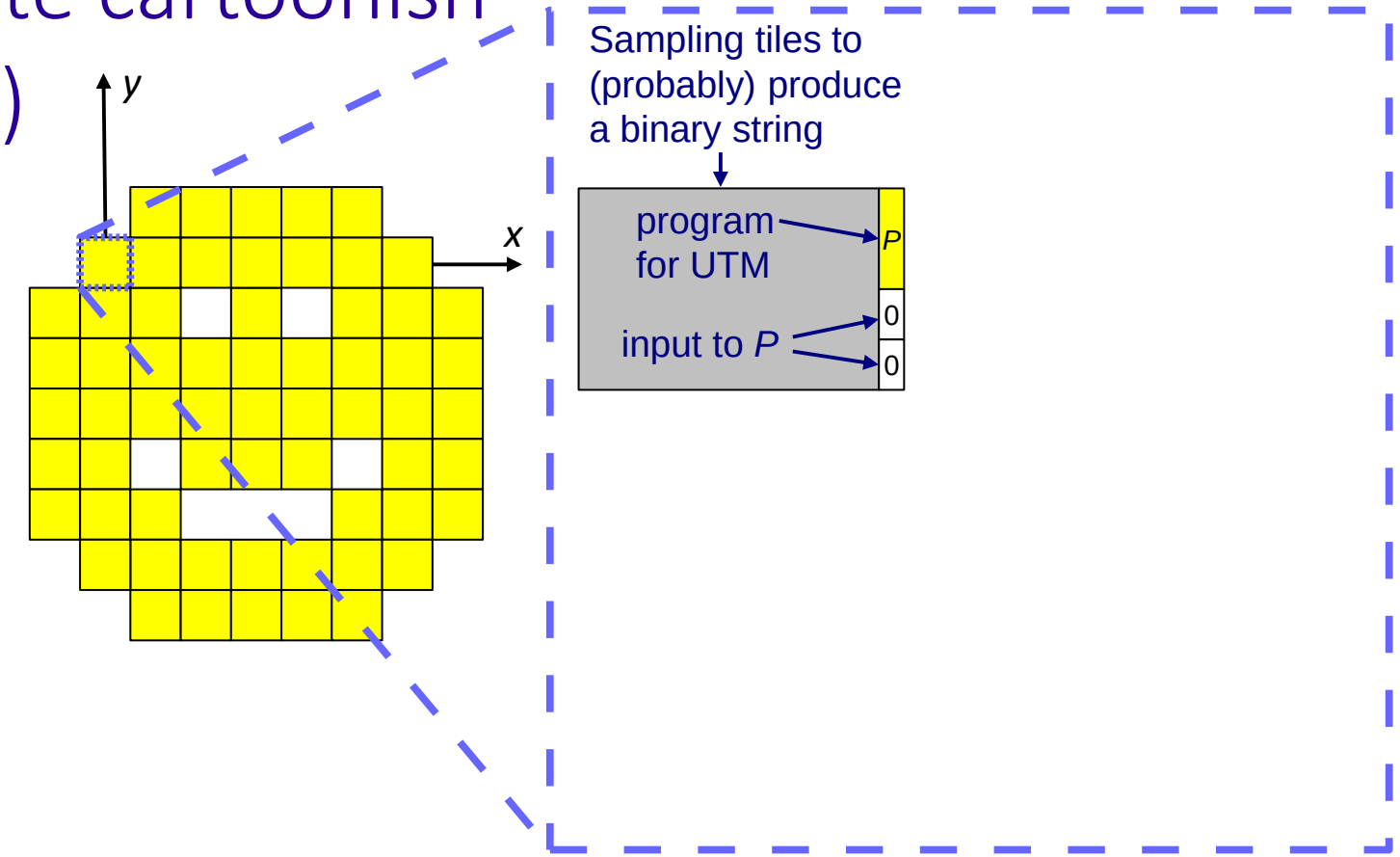

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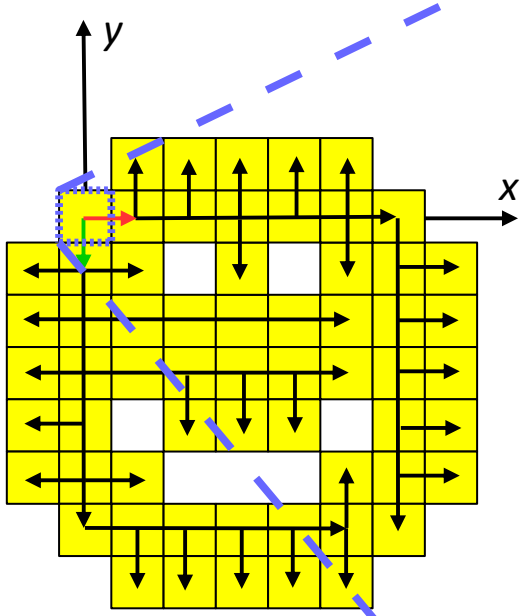
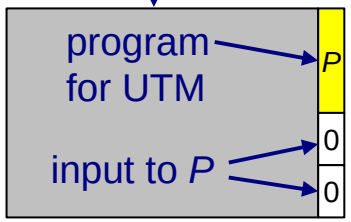


Sampling tiles to (probably) produce a binary string



slight modification of how *P* "computes" shape *S*: *P*(*x*,*y*) computes **spanning tree** of *S*, outputs **children** of point (*x*,*y*)

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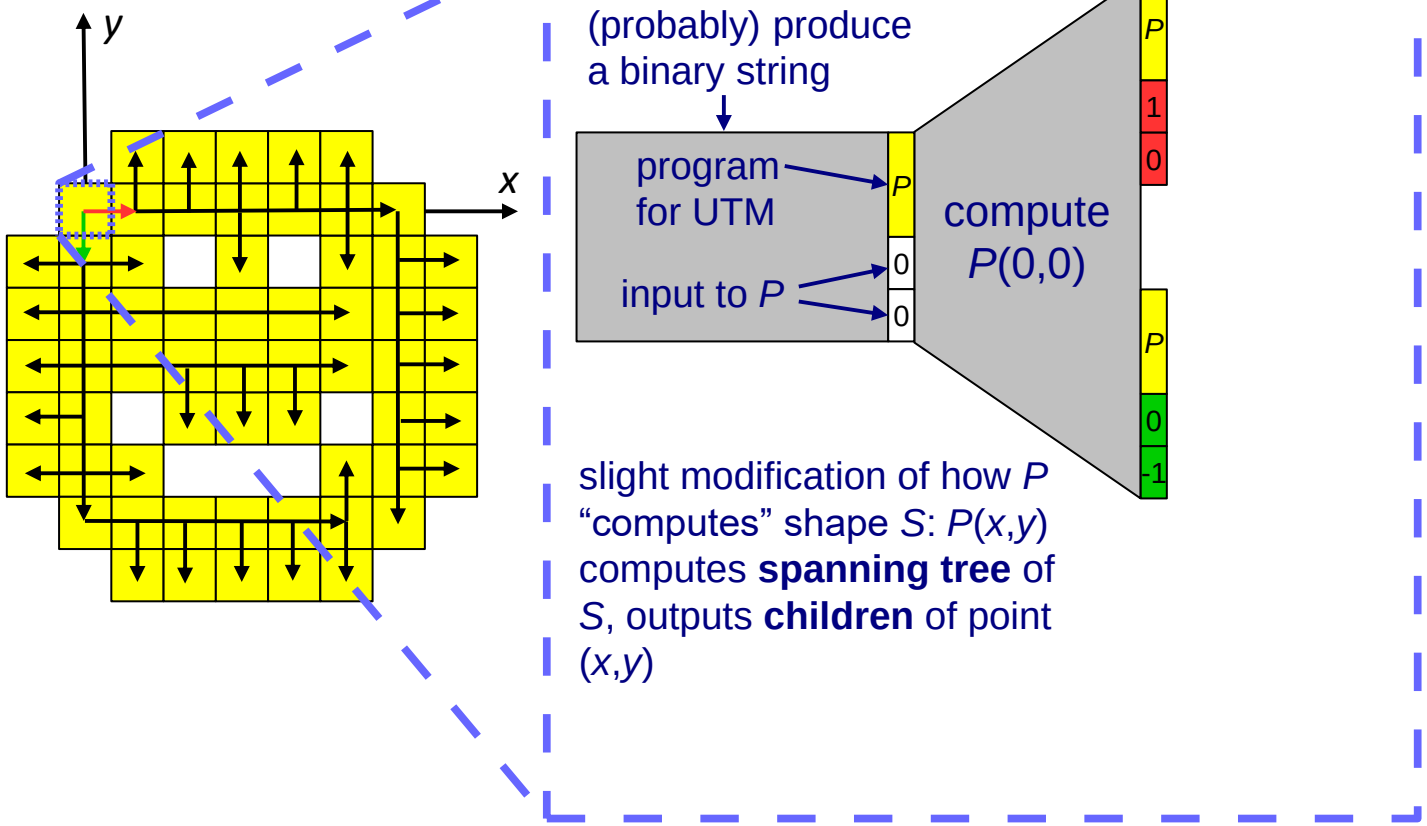

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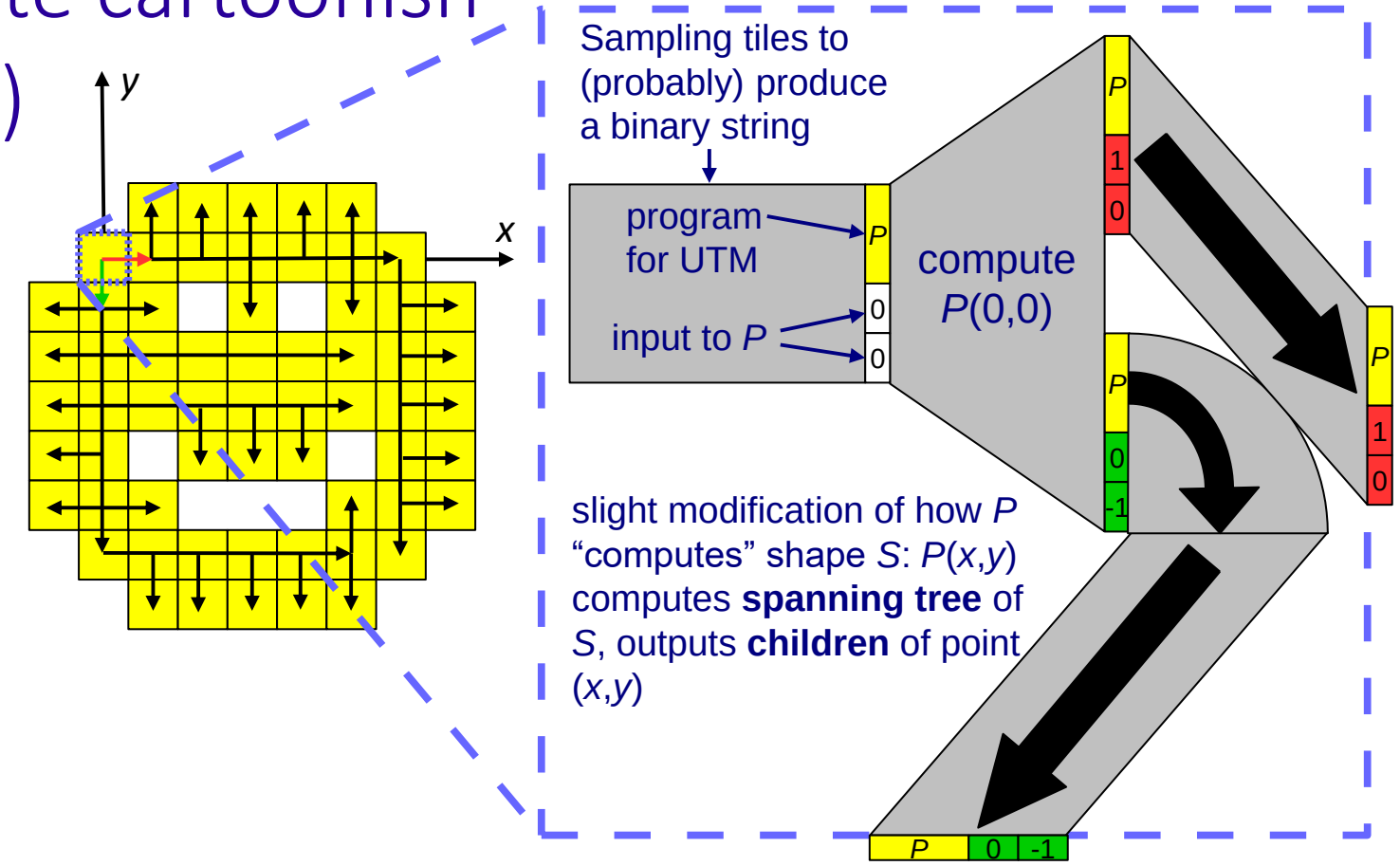

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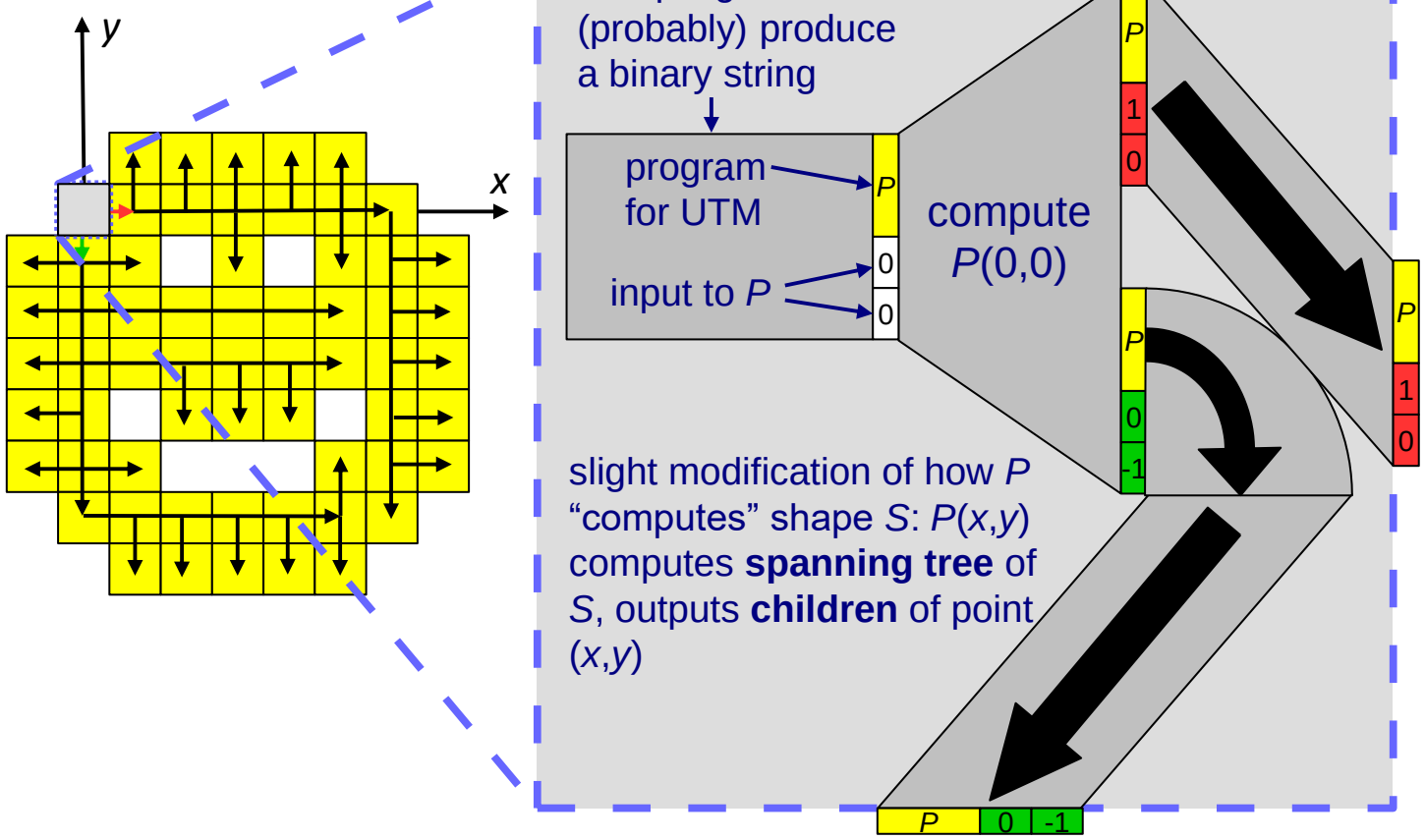

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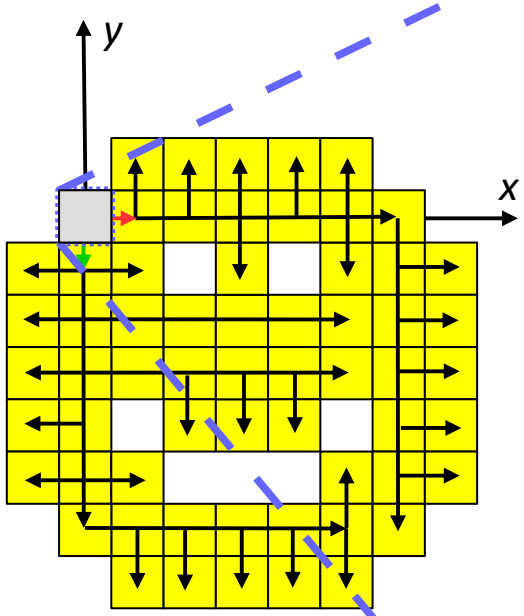

Sampling tiles to (probably) produce a binary string

program for UTM

input to *P*

compute *P*(0,0)

slight modification of how *P* "computes" shape *S*: *P*(*x*,*y*) computes **spanning tree** of *S*, outputs **children** of point (*x*,*y*)

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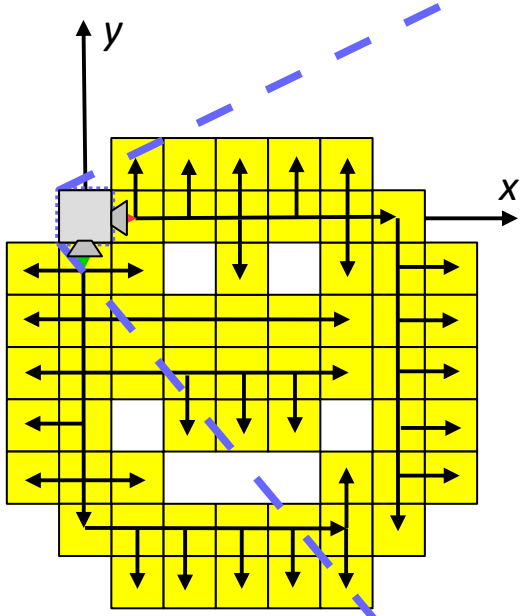

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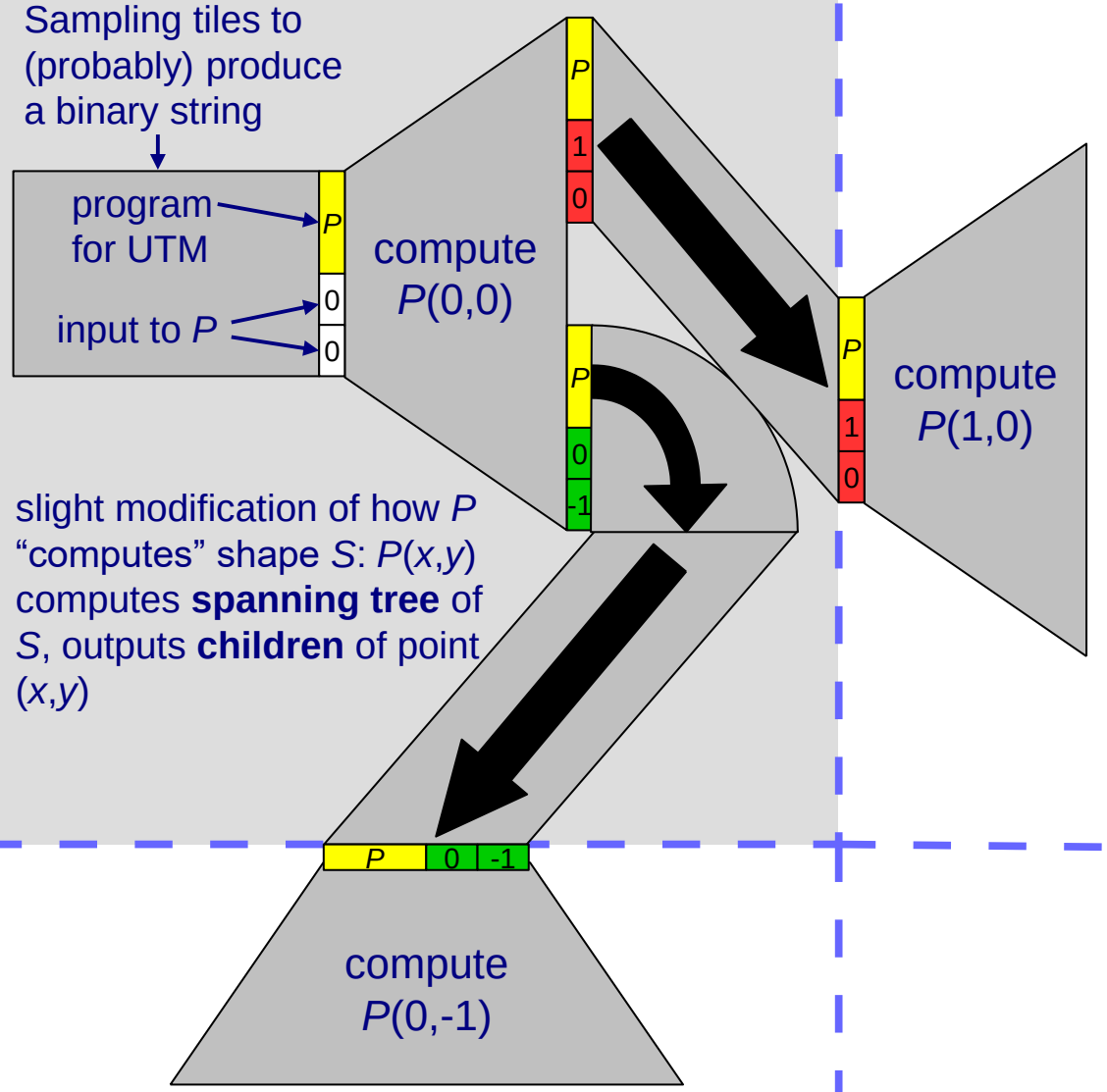

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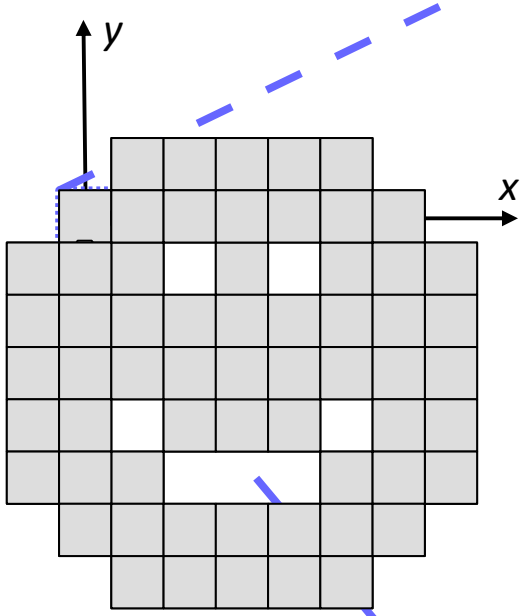

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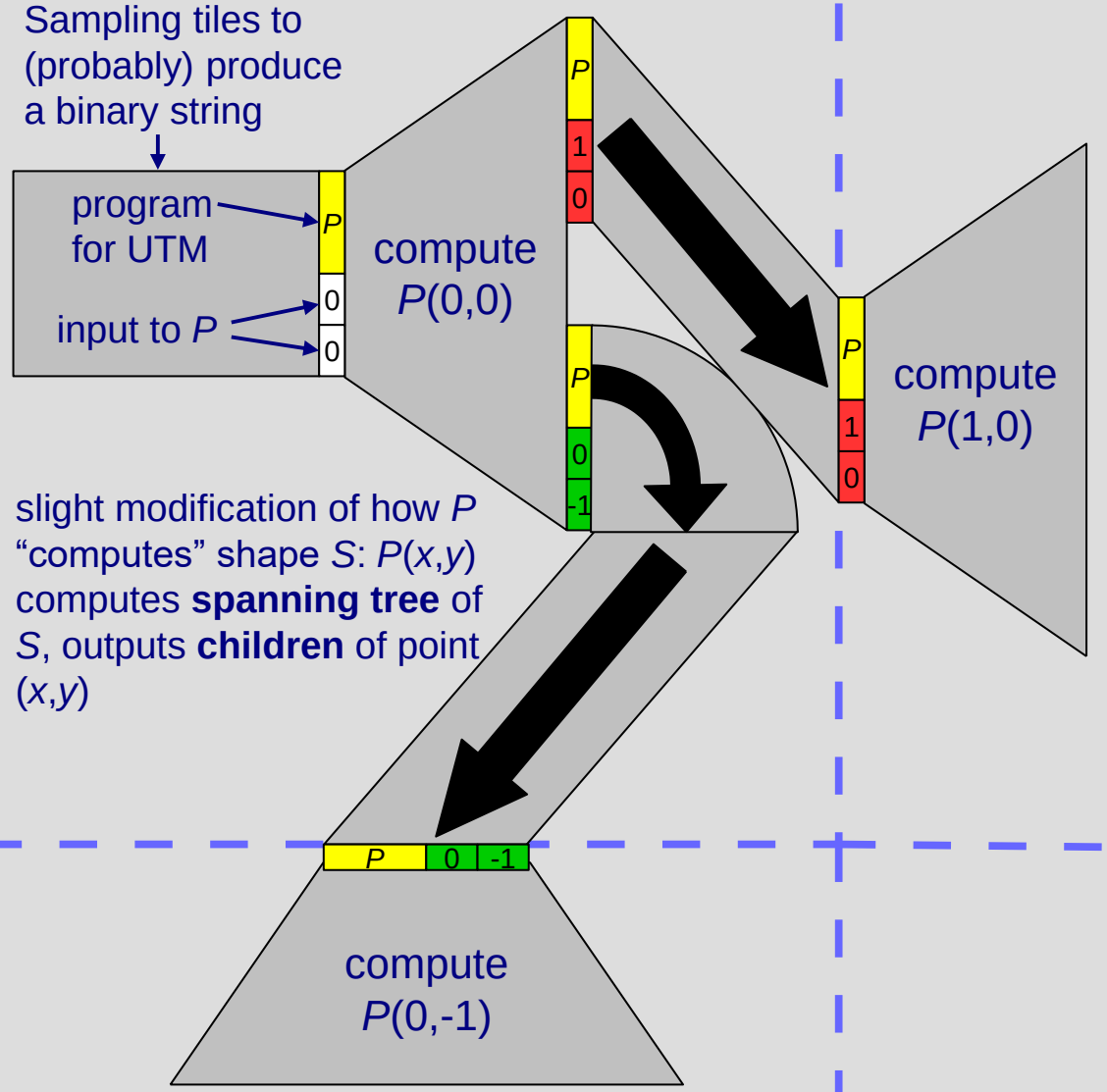

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Universal self-assembling molecules

A **fixed** set of tile types can assemble *any* finite (scaled) shape (with high probability) by mixing them in the right concentrations.

[Doty, *Randomized self-assembly for exact shapes*, SICOMP 2010, FOCS 2009]

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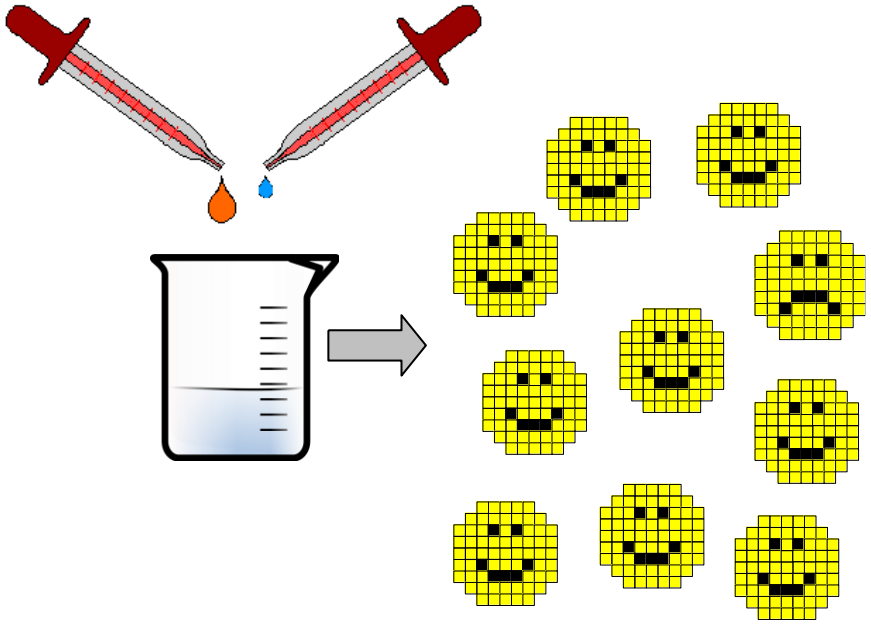

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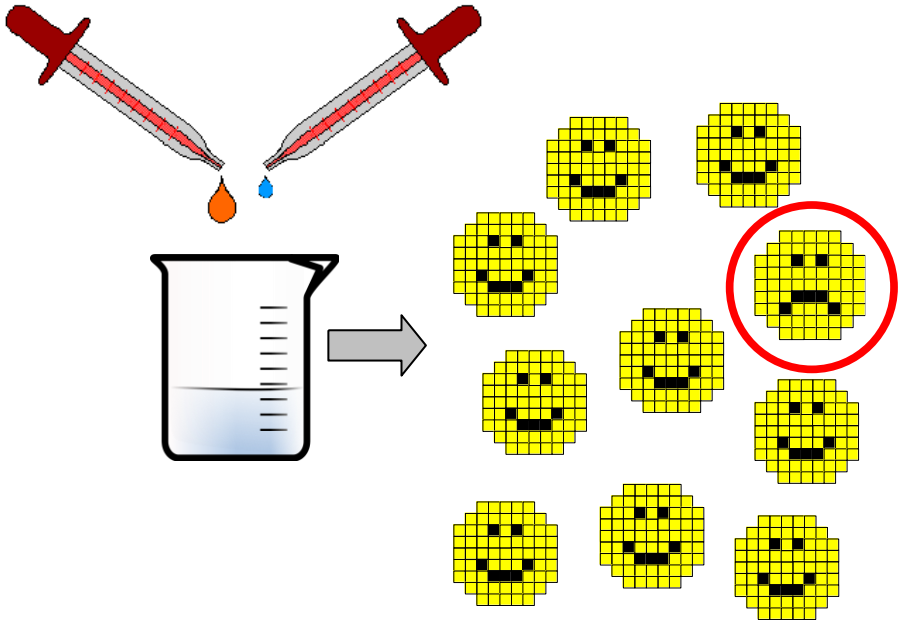

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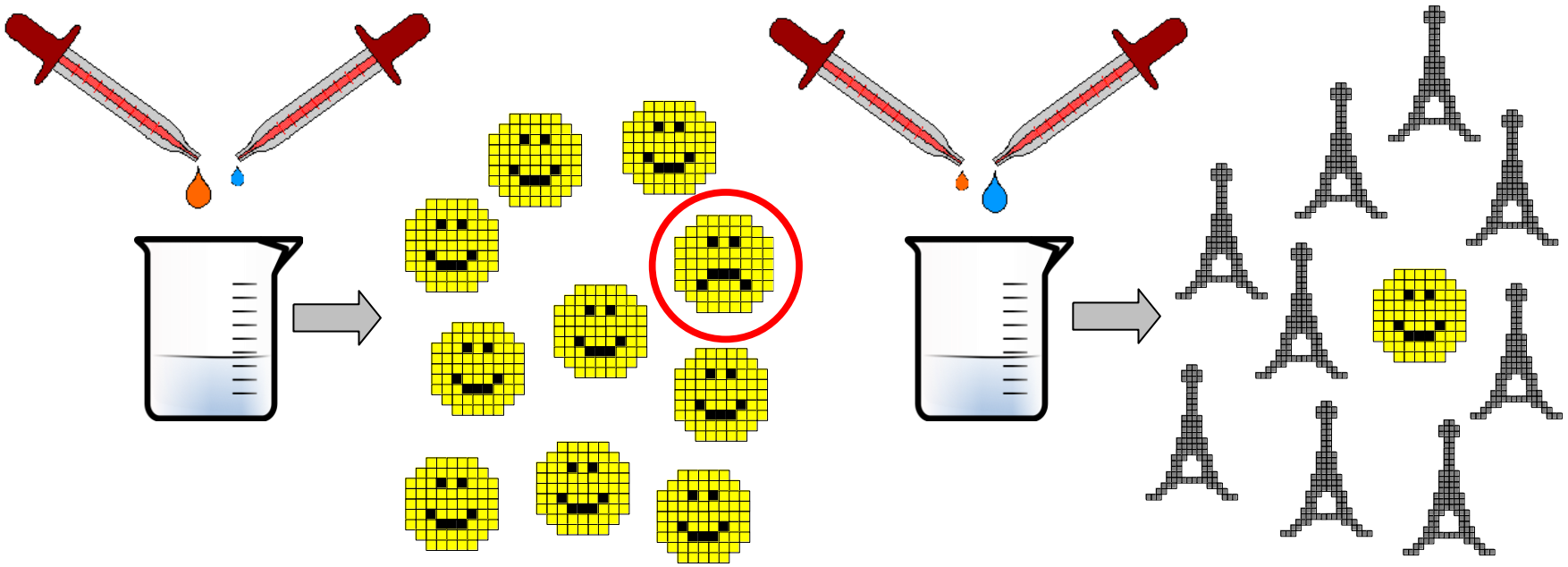

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Other plausible modifications of aTAM model that can reduce tile complexity

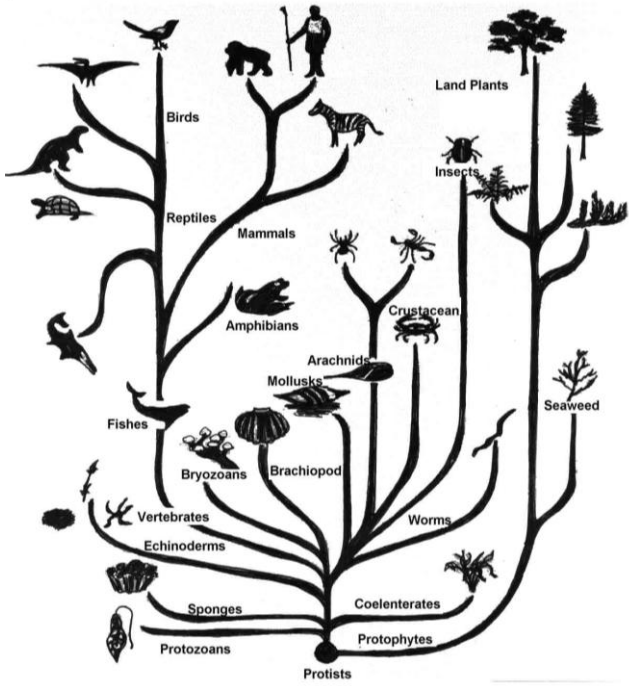
- staged self-assembly:
  - https://doi.org/10.1007/s11047-008-9073-0
- temperature programming:
  - https://dl.acm.org/doi/10.5555/1109557.1109620

The power of nondeterminism in self-assembly

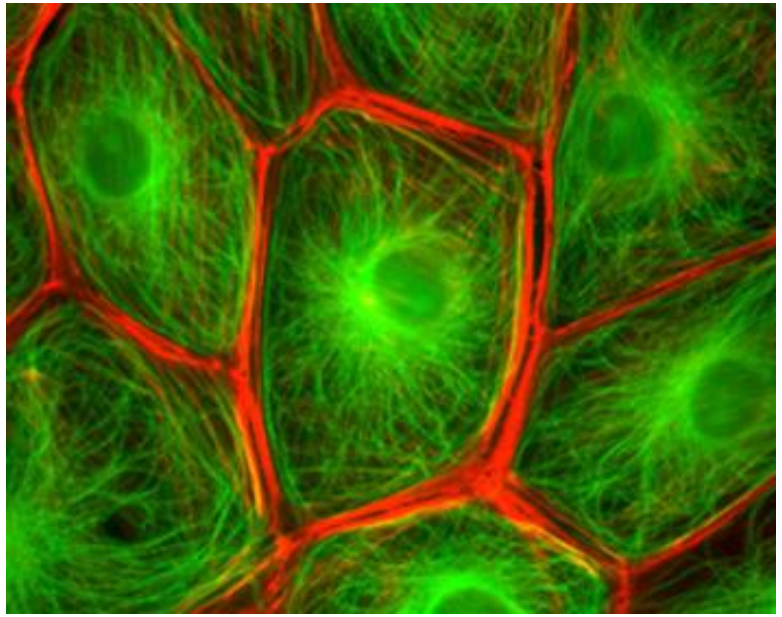
Can nondeterminism help to self-assemble shapes?

Nondeterminism in Biology

Genetic mutation



Cytoskeleton formation



Nondeterminism can allow complex structures to be created from a compact encoding.

Nondeterminism in Computer Science

Algorithm types:

Nondeterminism in Computer Science

Algorithm types:

Deterministic: entire computation uniquely determined by input

Nondeterminism in Computer Science

Algorithm types:

Randomized:
flips coins; realistic

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Nondeterminism in Computer Science

Algorithm types:

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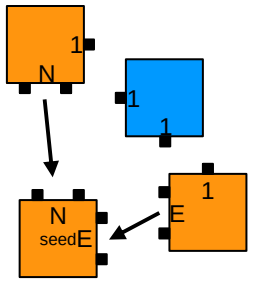
Trivially nondeterministic ("pseudodeterministic"):
flips coins, but *final output* independent of flip results

Deterministic: entire computation uniquely determined by input

Nondeterminism in Self-Assembly

Perhaps:



≥ 2 potential binding sites

Nondeterminism in Self-Assembly

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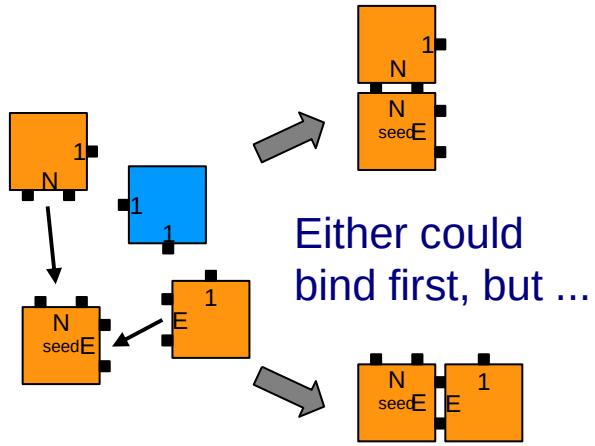

Either could bind first, but ...

Nondeterminism in Self-Assembly

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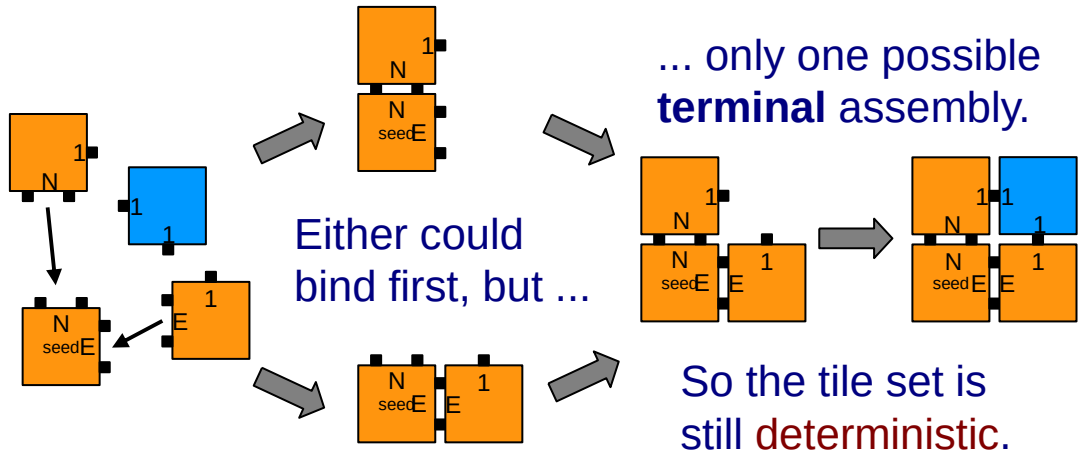

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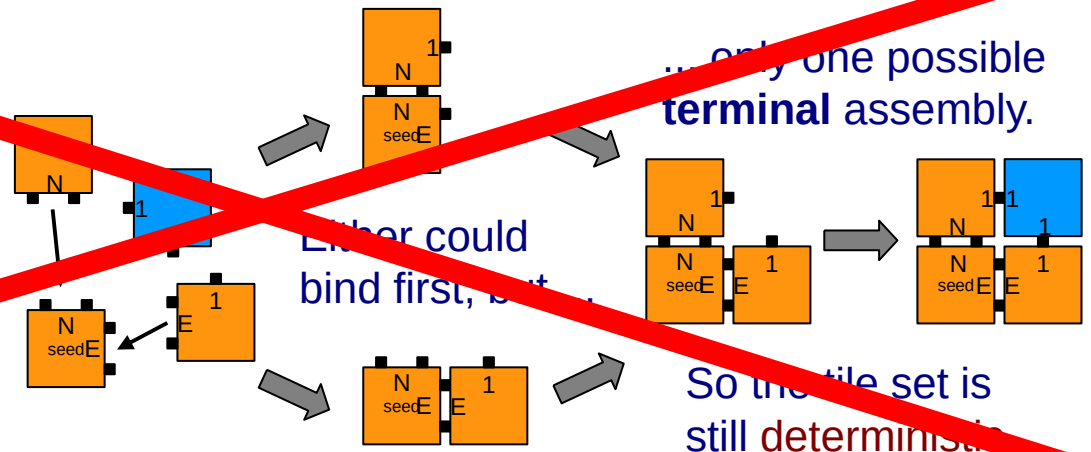
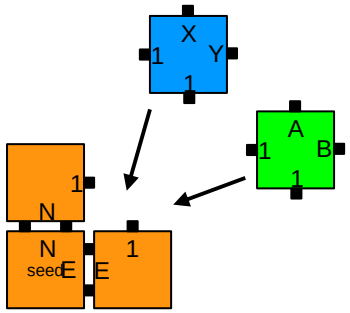


More meaningful:

at a *single* binding site, ≥ 2 tile types attachable



If tile types compete ...

Nondeterminism in Self-Assembly

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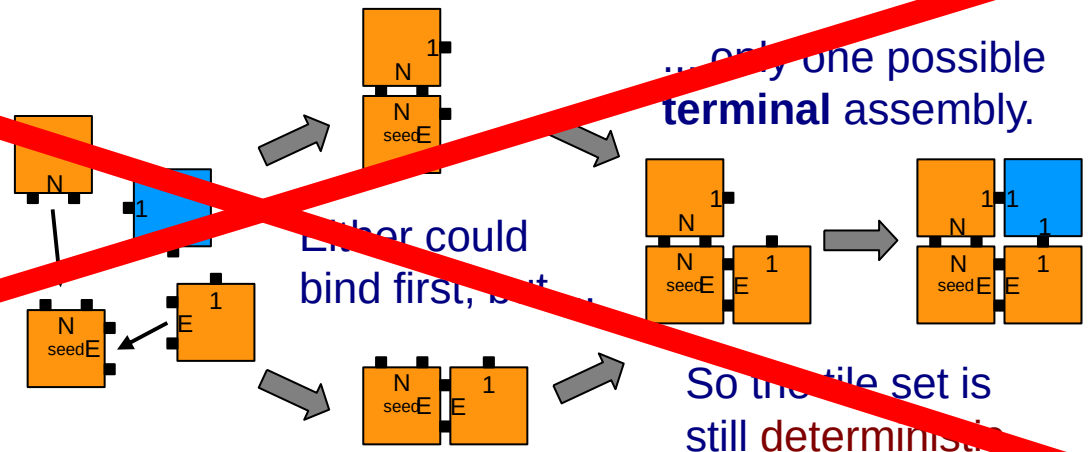

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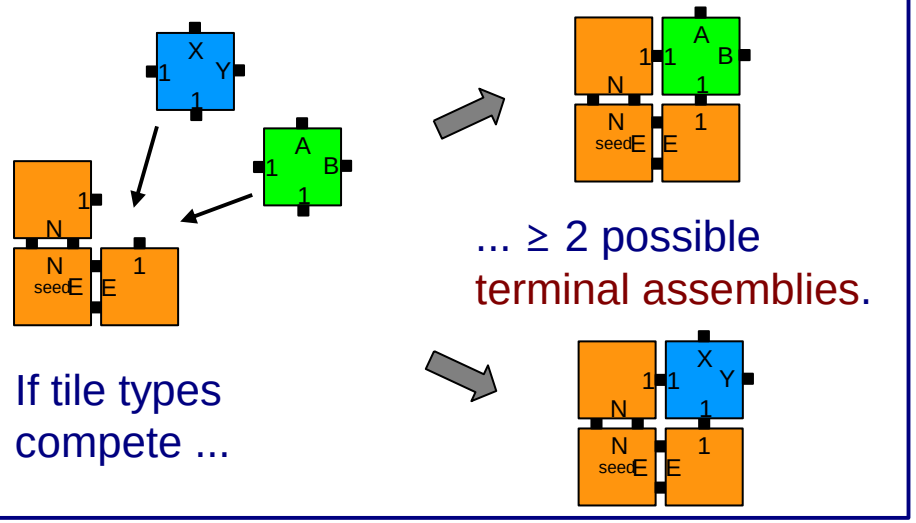

Nondeterminism in Self-Assembly

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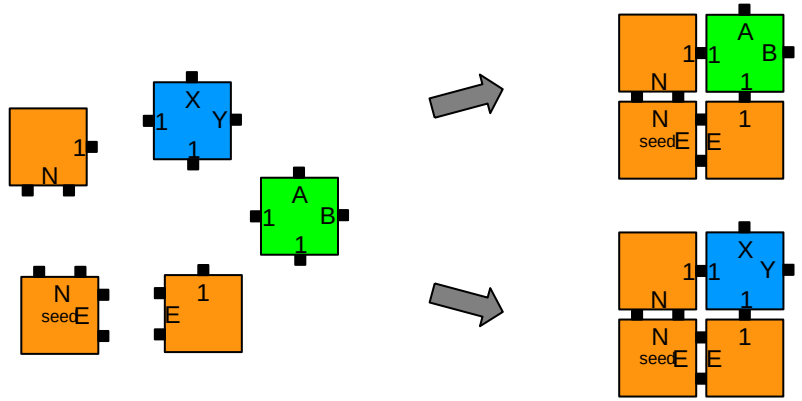
- This tile set has multiple terminal assemblies, *but* they all have the same shape.



- The tile set **self-assembles** a 2 x 2 square.

Power of Nondeterminism

Question: Let *S* be a finite shape self-assembled by some nondeterministic tile set. Does some deterministic tile set also self-assemble *S*?

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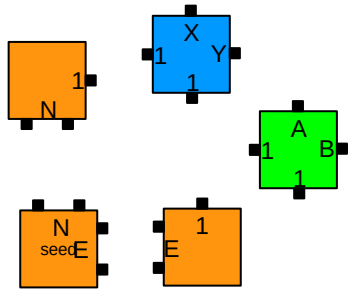

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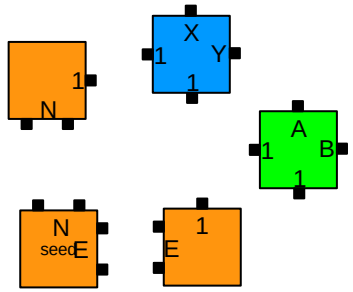
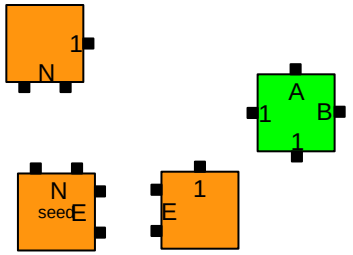


... to this deterministic tile set that self-assembles the same shape.

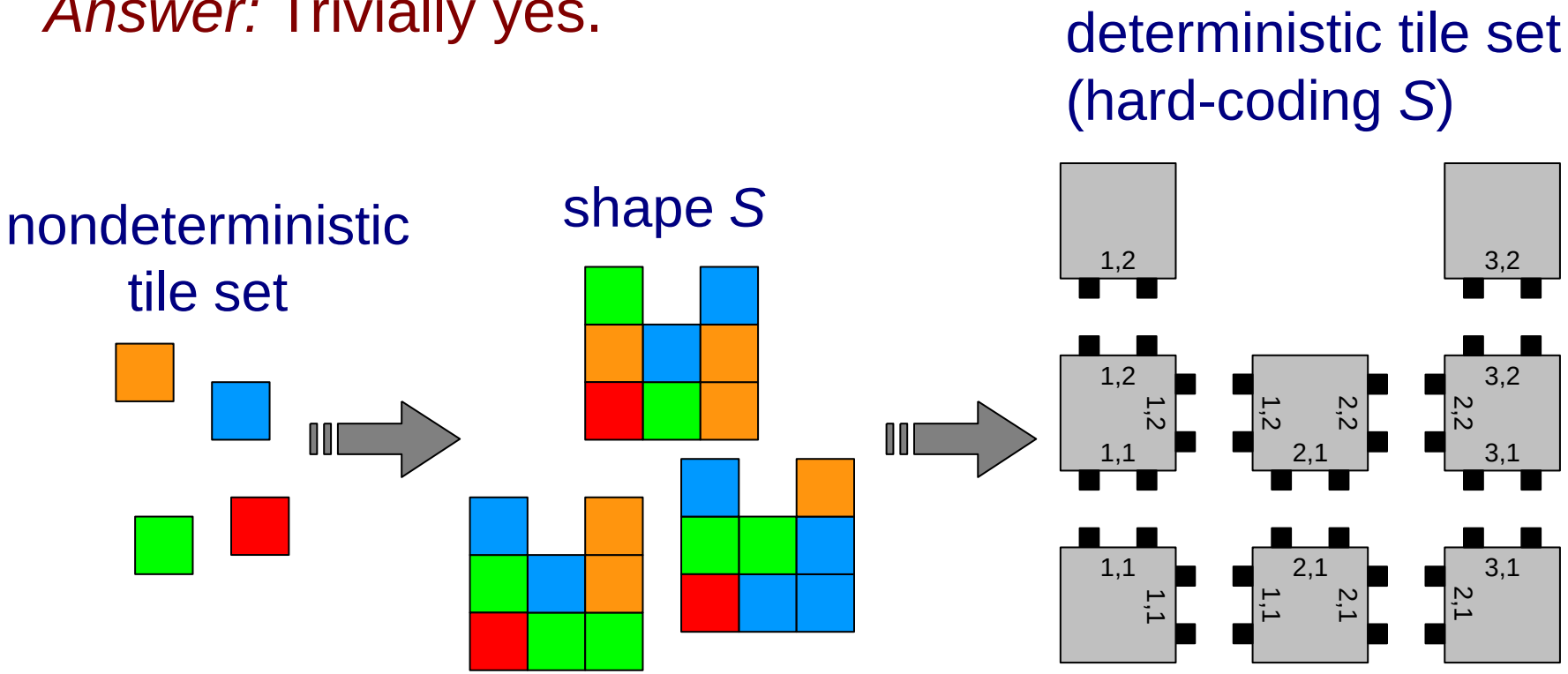


In general???

Power of Nondeterminism

Question: Let *S* be a finite shape self-assembled by some nondeterministic tile set. Does some deterministic tile set also self-assemble *S*?

Answer: Trivially yes.



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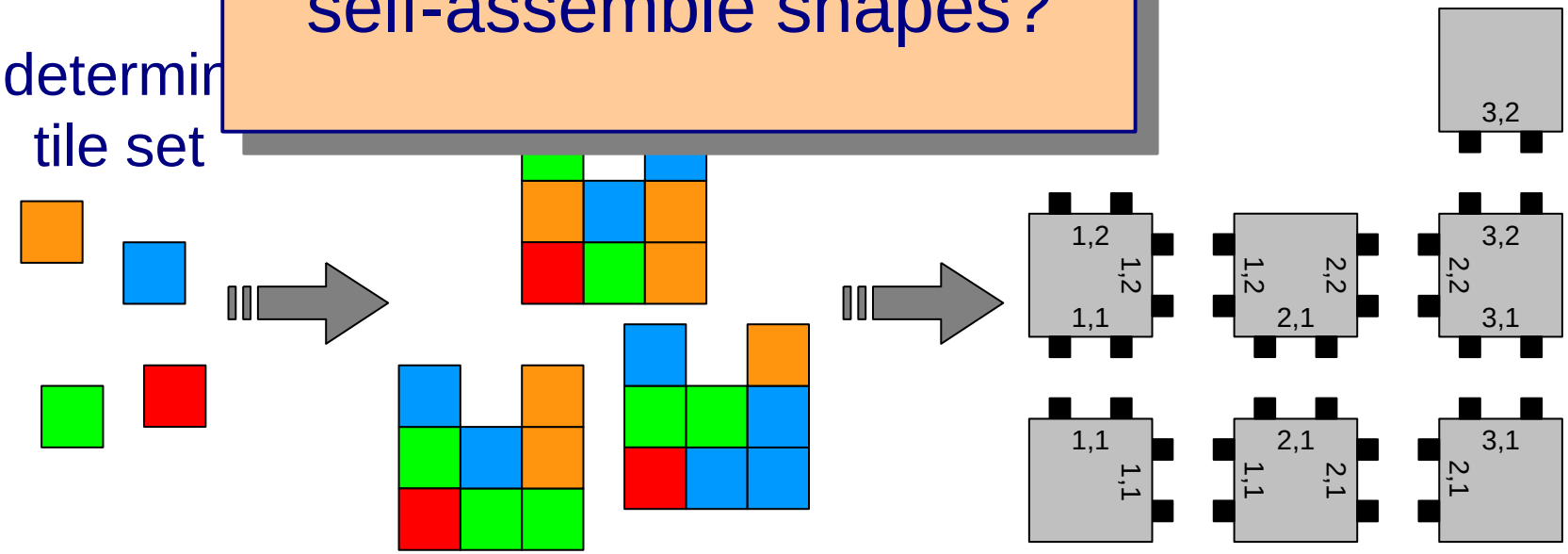
Is there **some** way that nondeterminism helps to self-assemble shapes?

Answer: T rministic tile set
d-coding *S*)

nondetermin
tile set



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Remainder of talk

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Given: finite shape *S*

Find: size of smallest tile system that self-assembles *S*

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if true, would imply MINDETTILESET = MINTILESET

Main Result

- We show: MINTILESET is **NP**$^{\mathbf{NP}}$-complete.
  a.k.a., Σ_2^P
- MINDETTILESET is **NP**-complete. (Adleman, Cheng, Goel, Huang, Kempe, Moisset de Espanés, Rothemund, *STOC* 2002)
- **NP** ≠ **NP**$^{\mathbf{NP}}$ ⇒ MINTILESET ≠ MINDETTILESET

Nondeterminism in Algorithms and Self-Assembly

Algorithm that flips coins but always produces same output

- coin flips **useless**

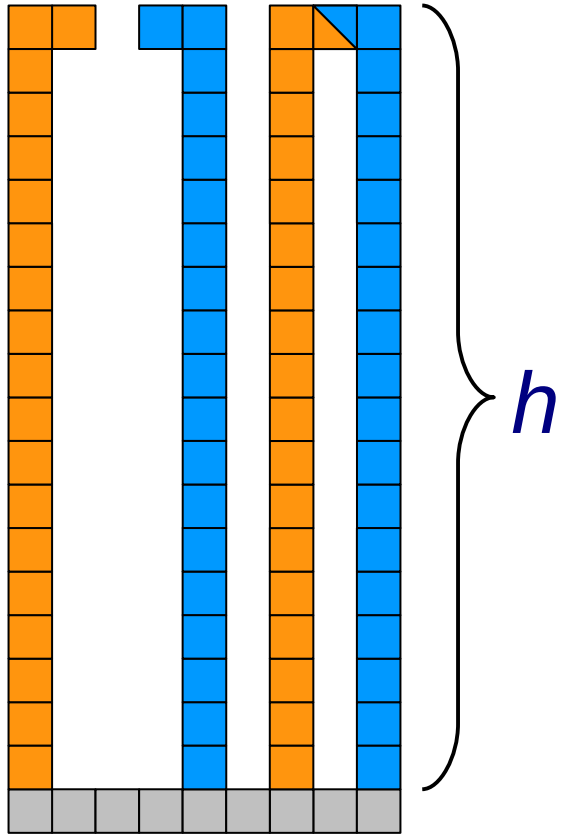
Tile set that flips coins but always produces same shape

- coin flips **useful**

But … finding smallest tile set is harder if it flips coins.

A Finite Shape for which Nondeterminism Affects Tile Complexity

- Smallest tile set: ≈ $2h$ tile types



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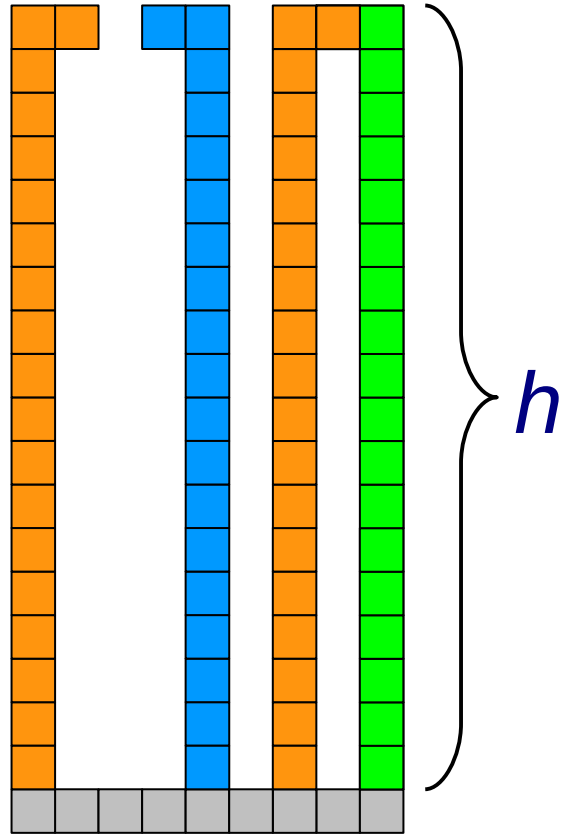

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in **NP**$^{\mathbf{NP}}$-hardness reduction, compete to assign bits to variable in Boolean formula

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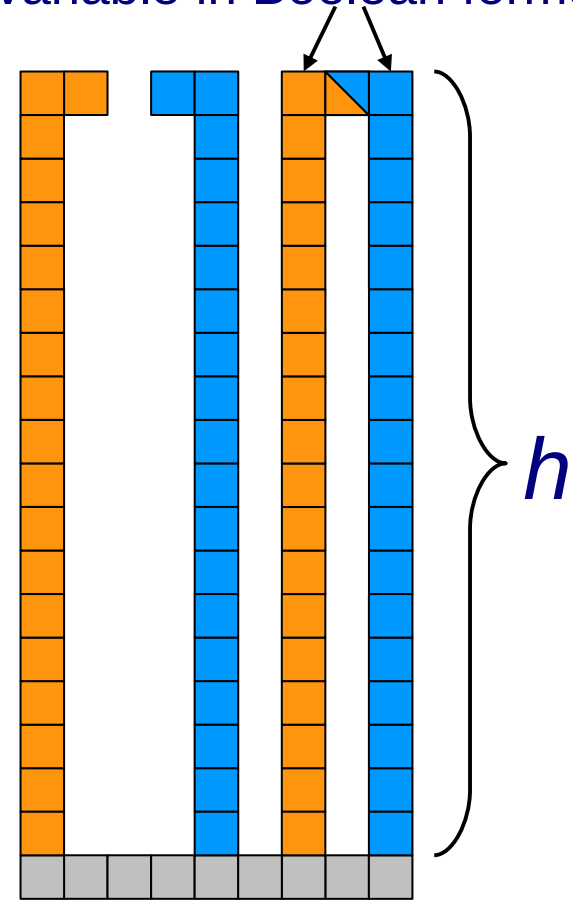

$\mathbf{NP^{NP}}$-hardness Reduction

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- **Reduction goal:** Given Φ, output shape S and integer c such that $(\exists x)(\forall y)\neg\Phi(x,y)$ holds if and only if some tile set of size at most c self-assembles S.

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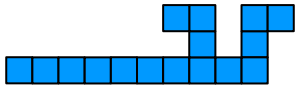

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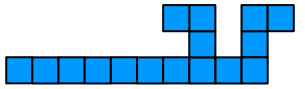

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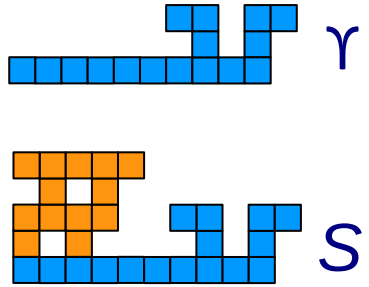

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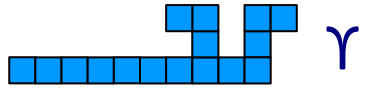

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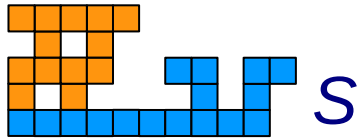

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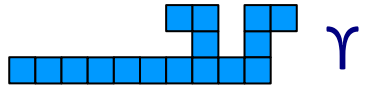

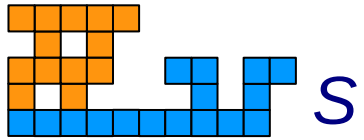

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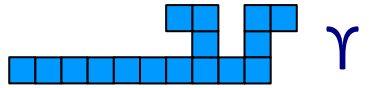
  - "Since $\Upsilon \subseteq S$," every tile set that assembles S contains T, so if tiles from T cannot be altered to assemble S then additional tiles are needed; i.e., S requires more than $c = |T|$ tile types.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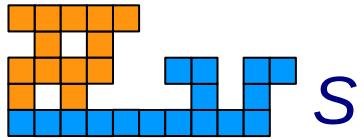
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C_3	SF	SF	ST	ST
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C_1	UF	UF	ST	ST
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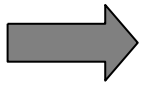
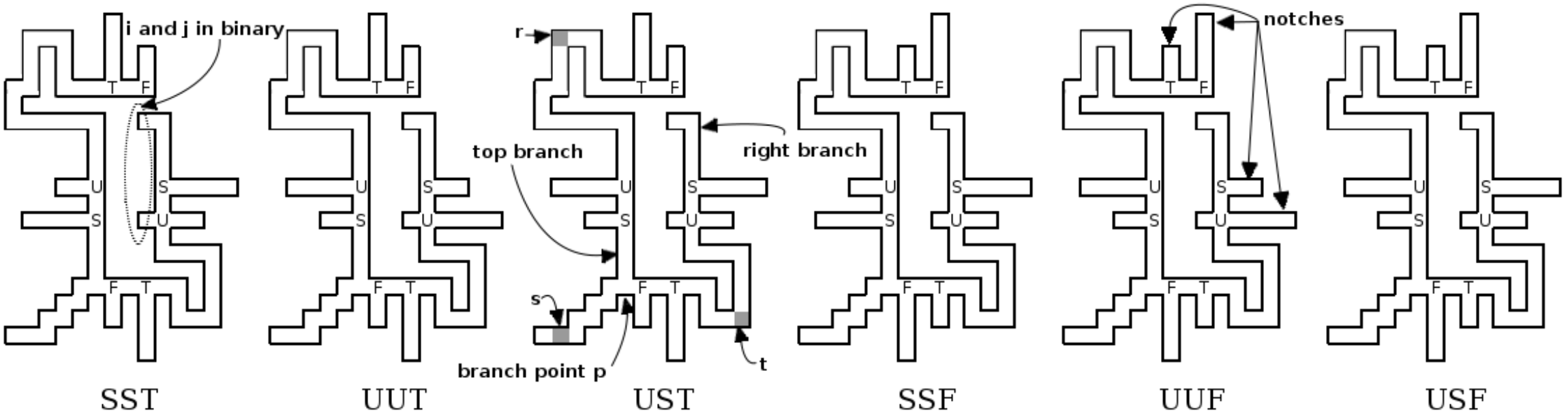


highlighting when C_i goes from unsatisfied (U) to satisfied (S)

C_3	USF	SSF	SST	SST
C_2	UUF	UUF	UUT	UST
C_1	UUF	UUF	UST	SST
	w_1	w_2	w_3	w_4

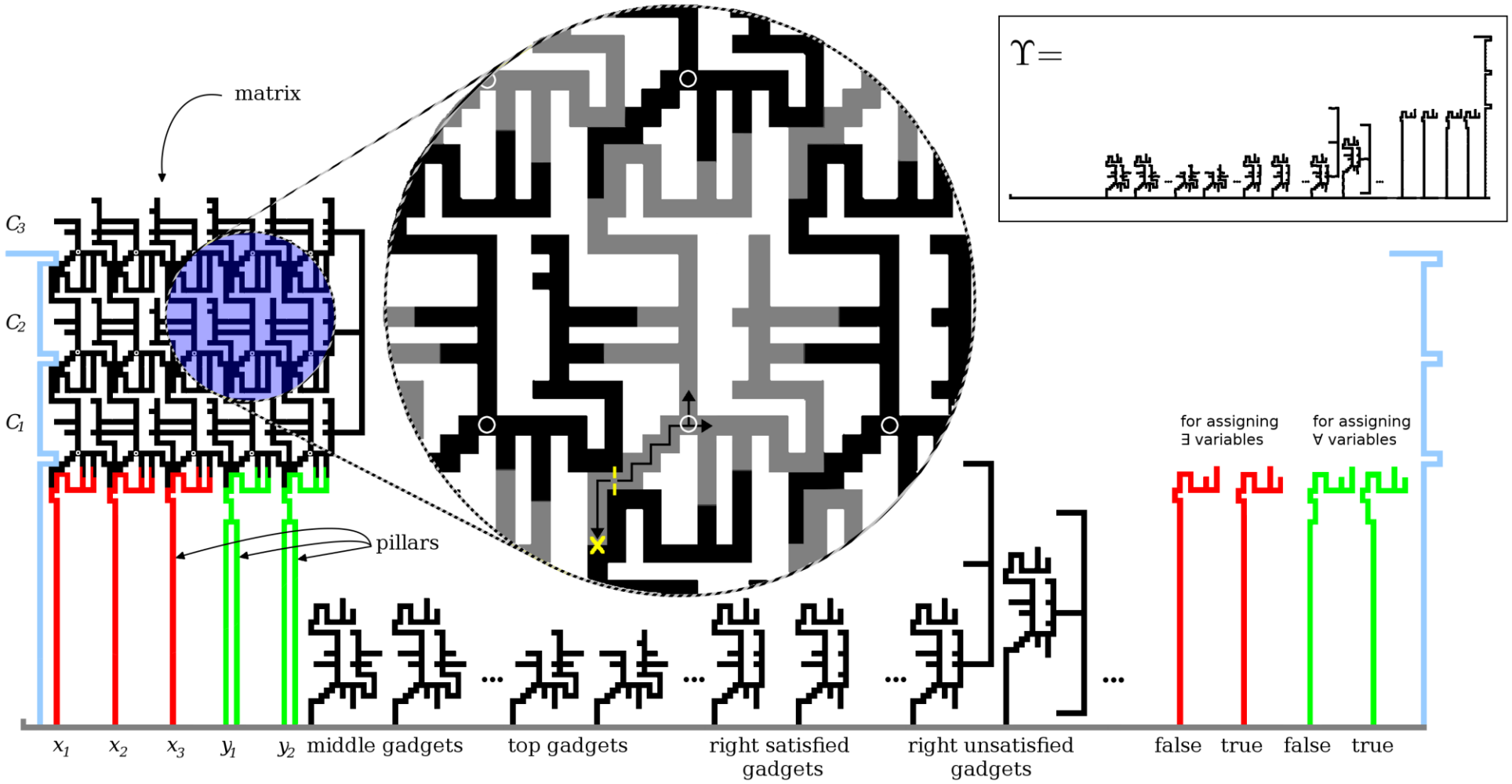
Gadgets (Adleman et al. 2002)



For each variable w_i and clause C_j, value of w_i = T/F and

SS_{ij} – C_j satisfied by a previous variable (w_k for $k < i$)
US_{ij} – C_j unsatisfied by previous variables but is satisfied by w_i
UU_{ij} – C_j unsatisfied by previous variables and by w_i

Shape *S*



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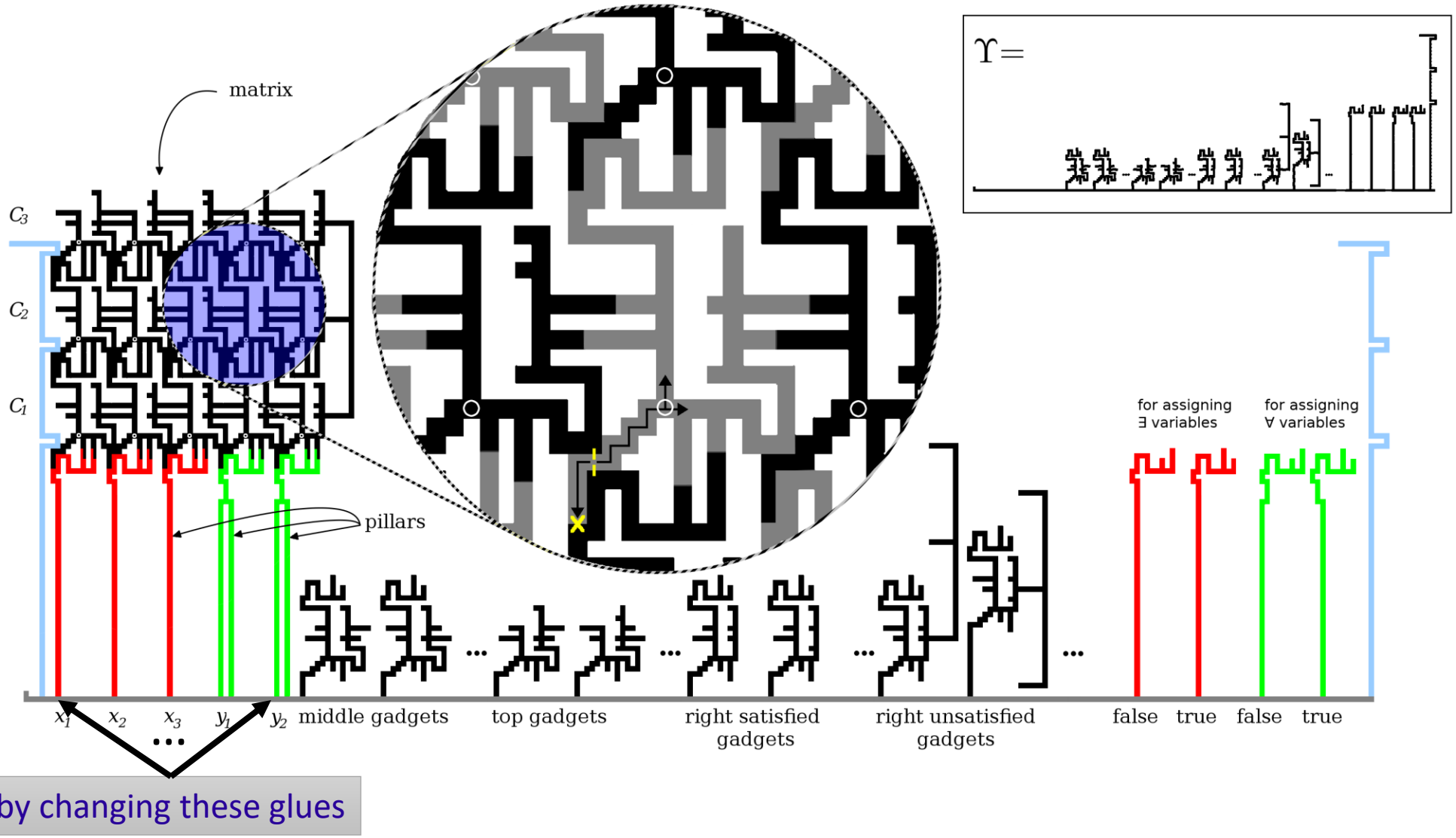

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  - deterministic case in **P**; likely not **NP**-hard by Mahaney's theorem (no sparse set is **NP**-hard unless **P**=**NP**)
- *Weak self-assembly* (pattern painting): paint some tile types “black”, and say “pattern assembled” is set of points with a black tile
  - *Minimum tile set problem:* uncomputable! (**NP**-complete with some restrictions: https://arxiv.org/abs/1404.0967)
  - *Power of nondeterminism:* is it possible to uniquely paint a pattern, but only by assembling more than one shape on which the pattern is painted?

Errors in algorithmic self-assembly

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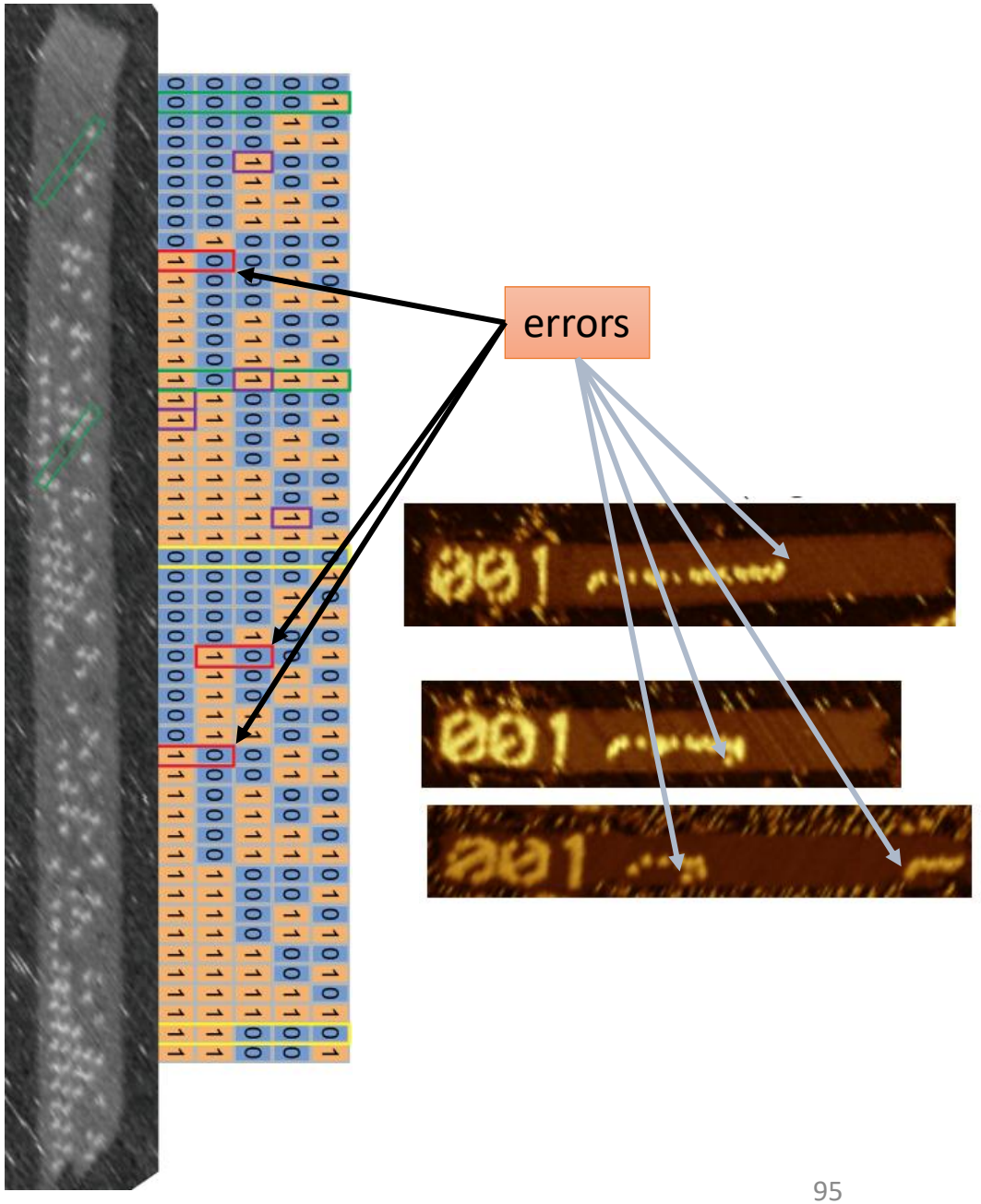

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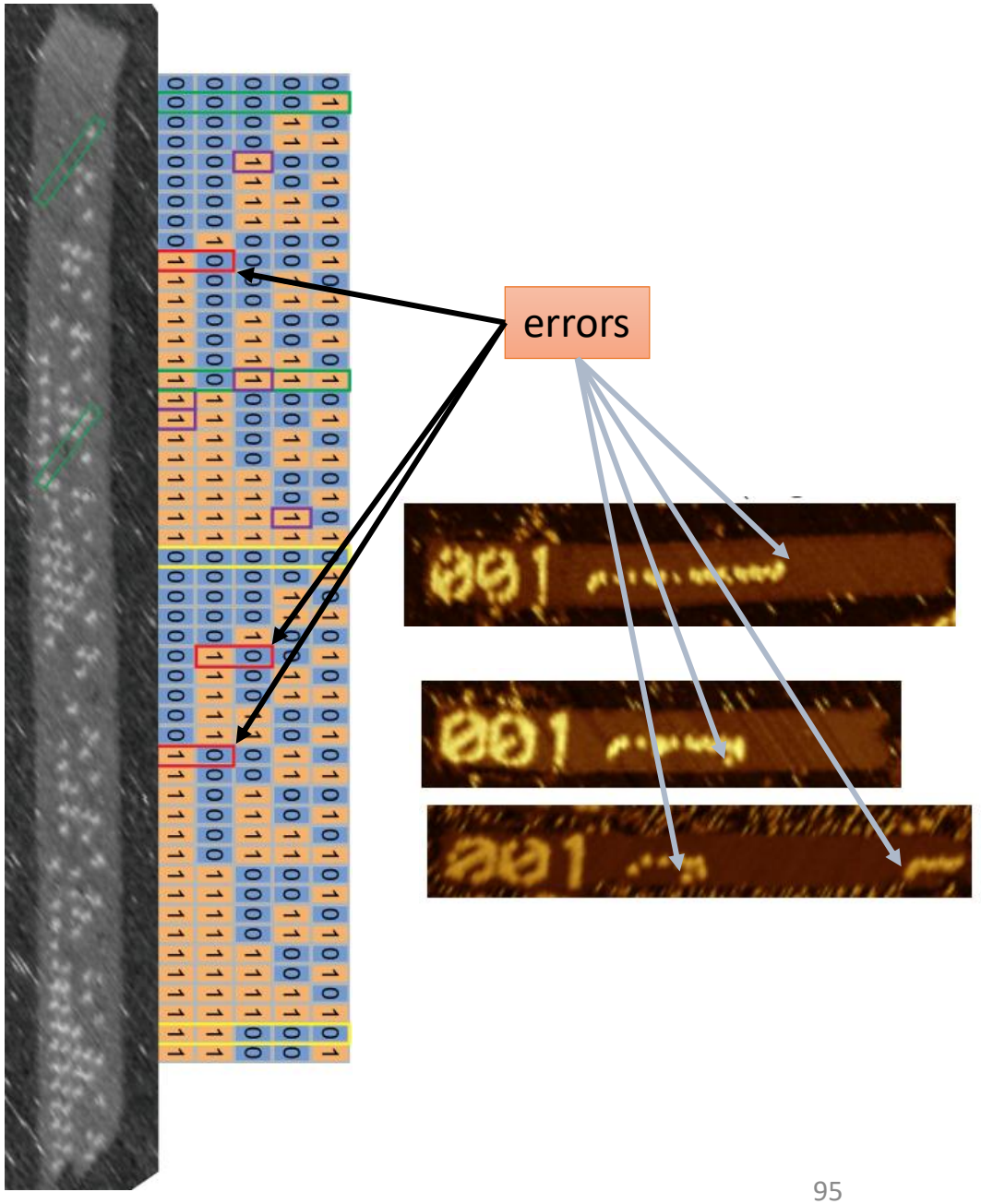

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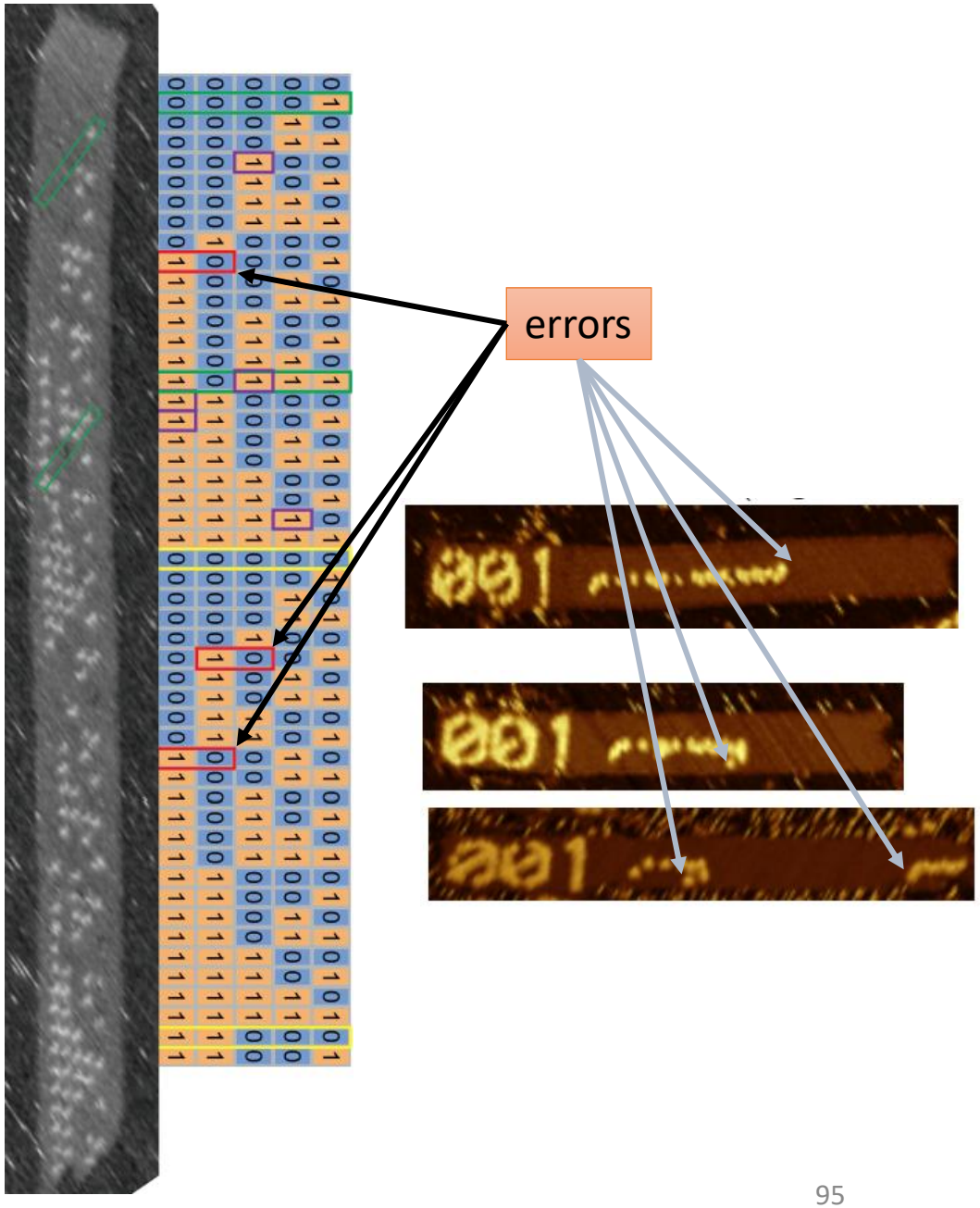
- kinetic Tile Assembly Model (kTAM); essential differences with aTAM:
  - tiles can detach
  - tiles can bind with strength 1



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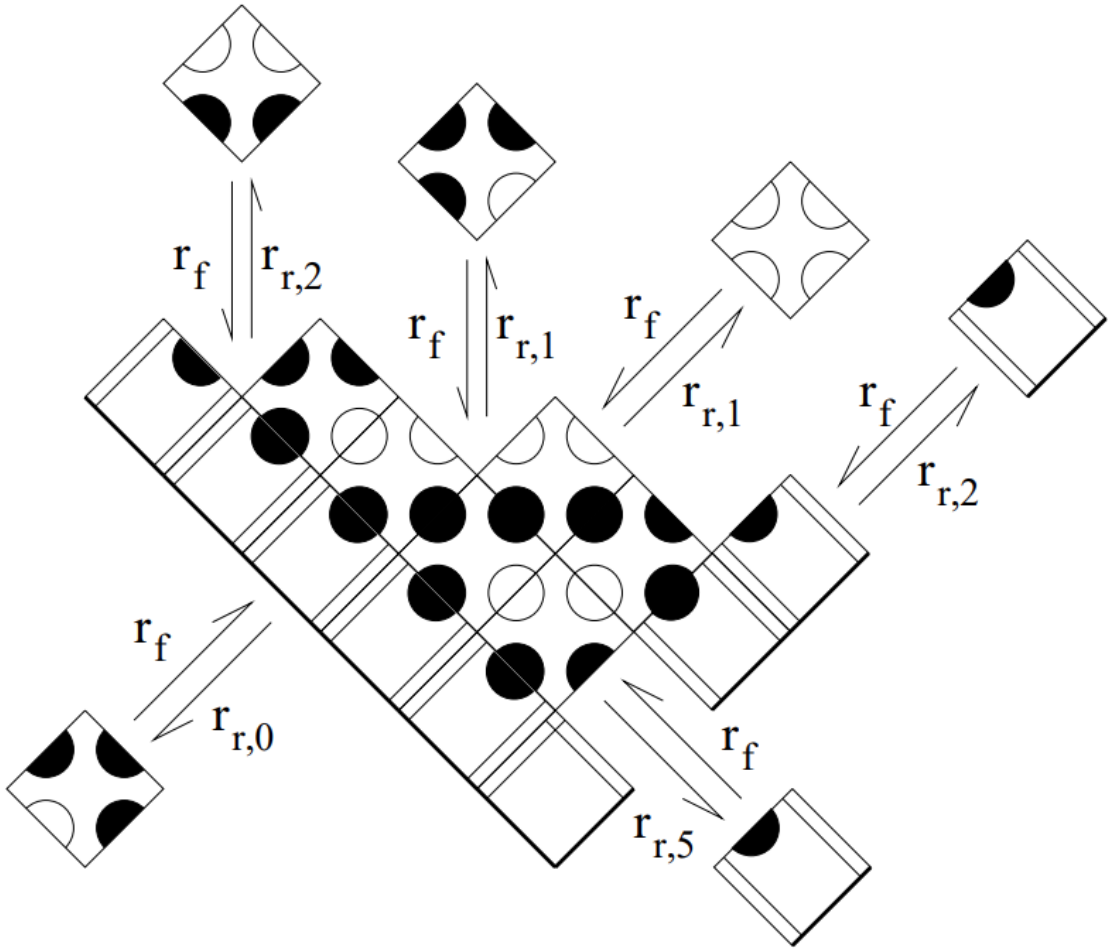

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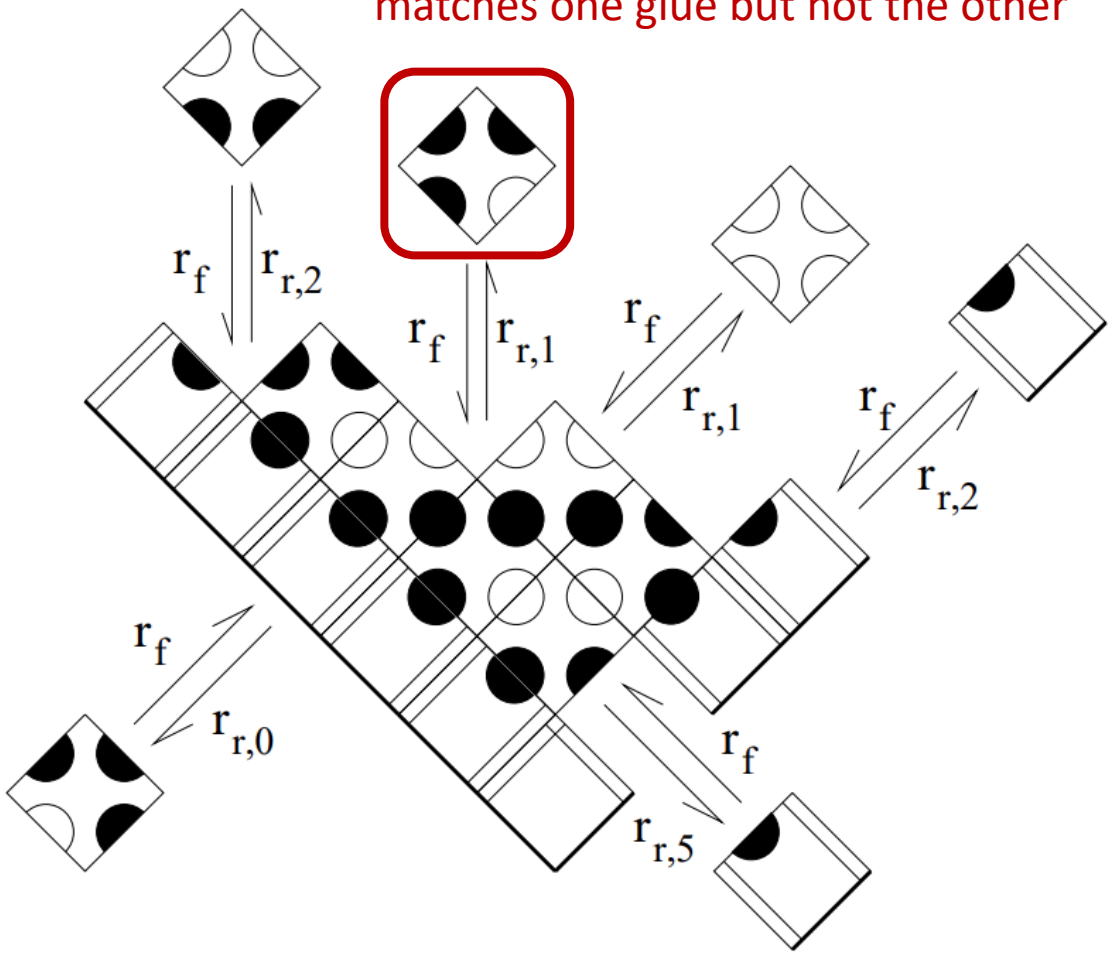

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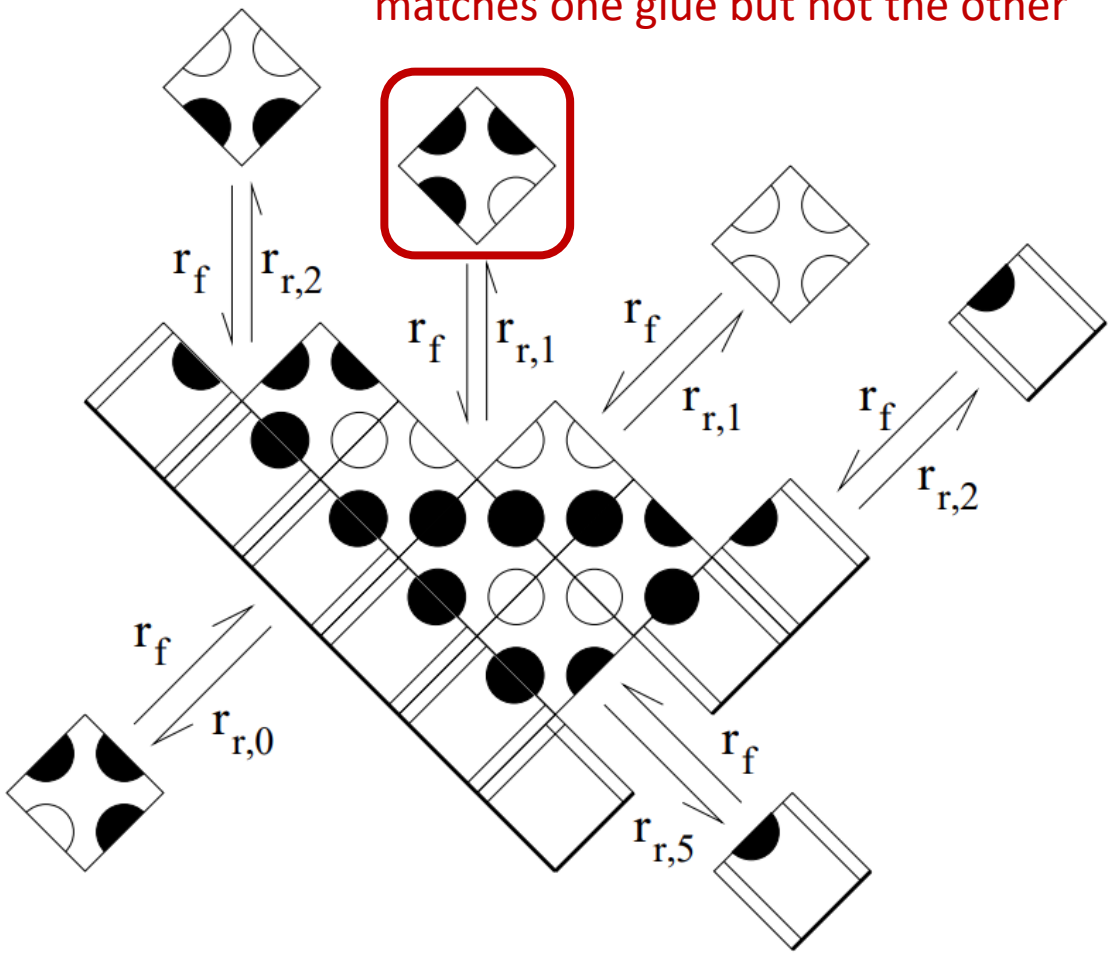

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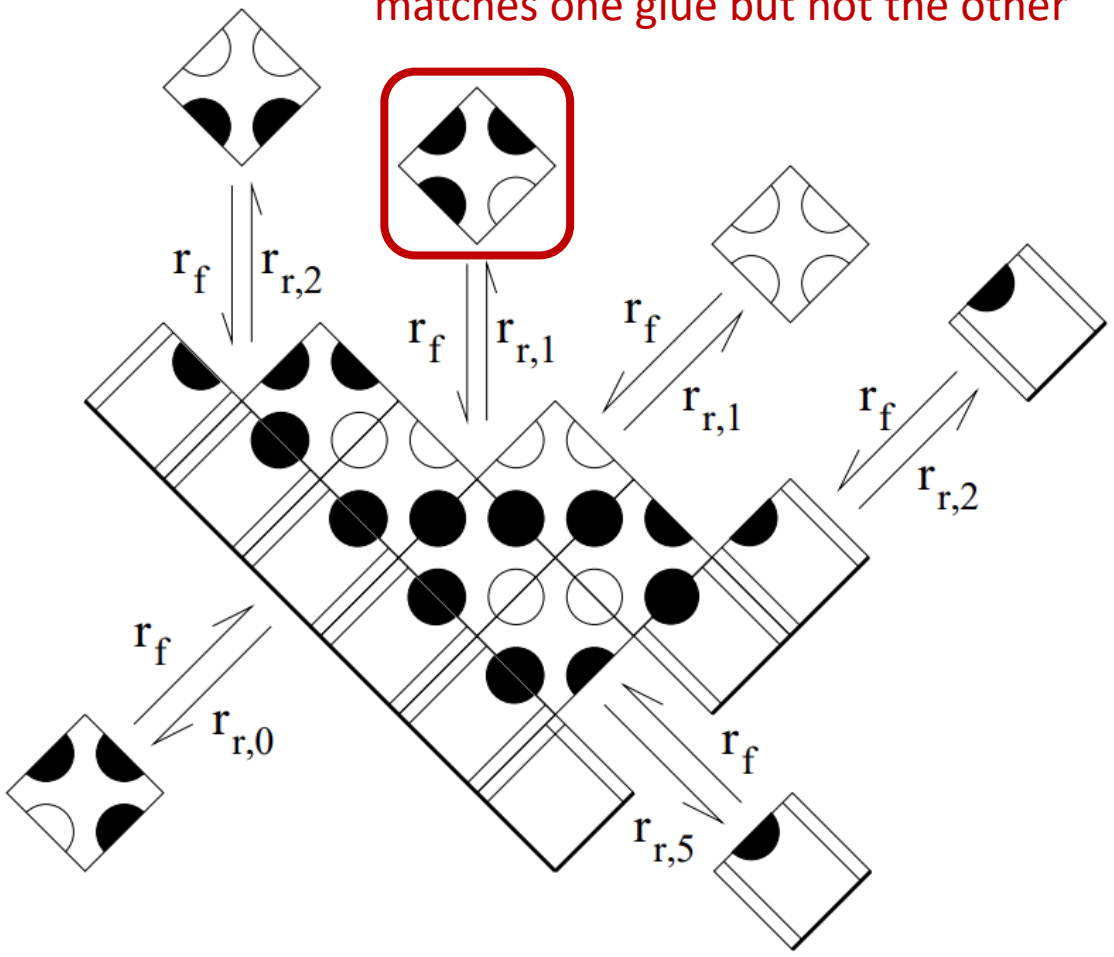

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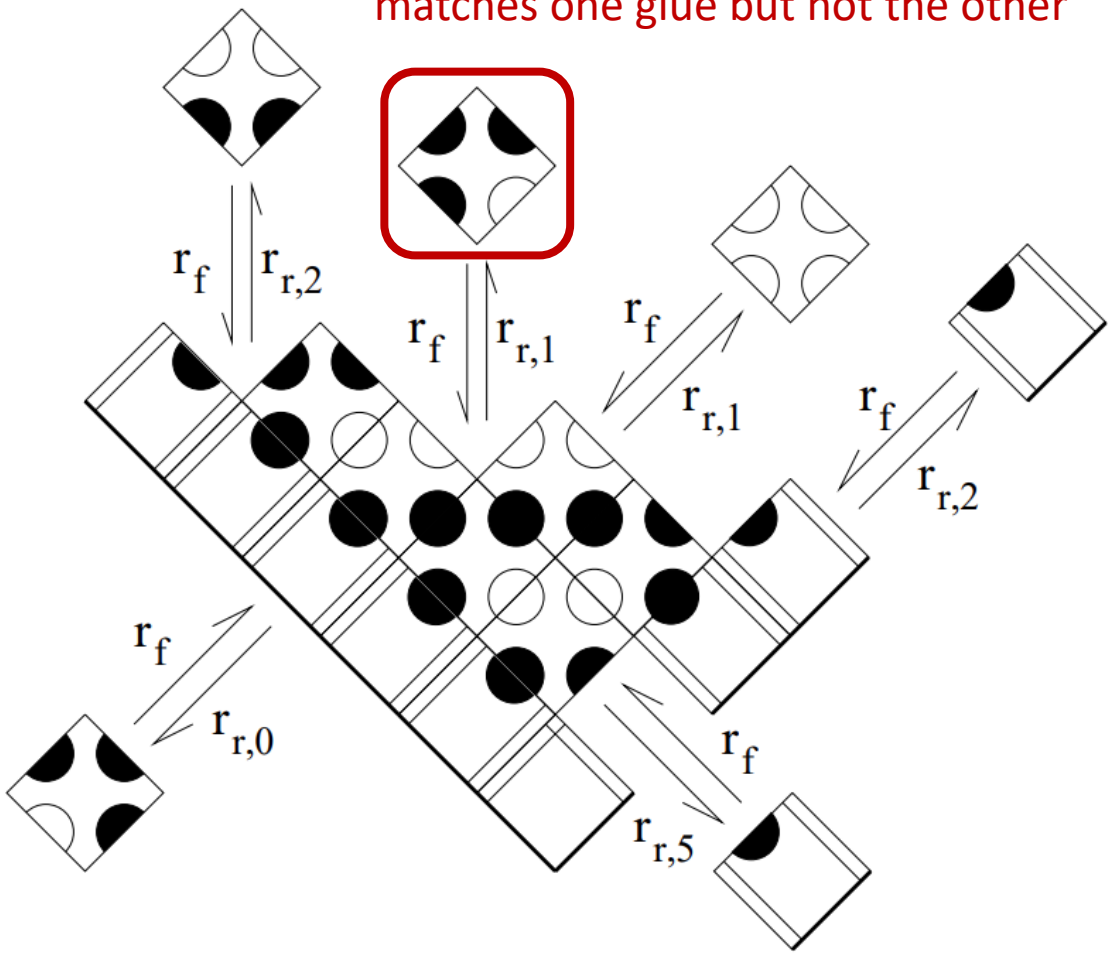

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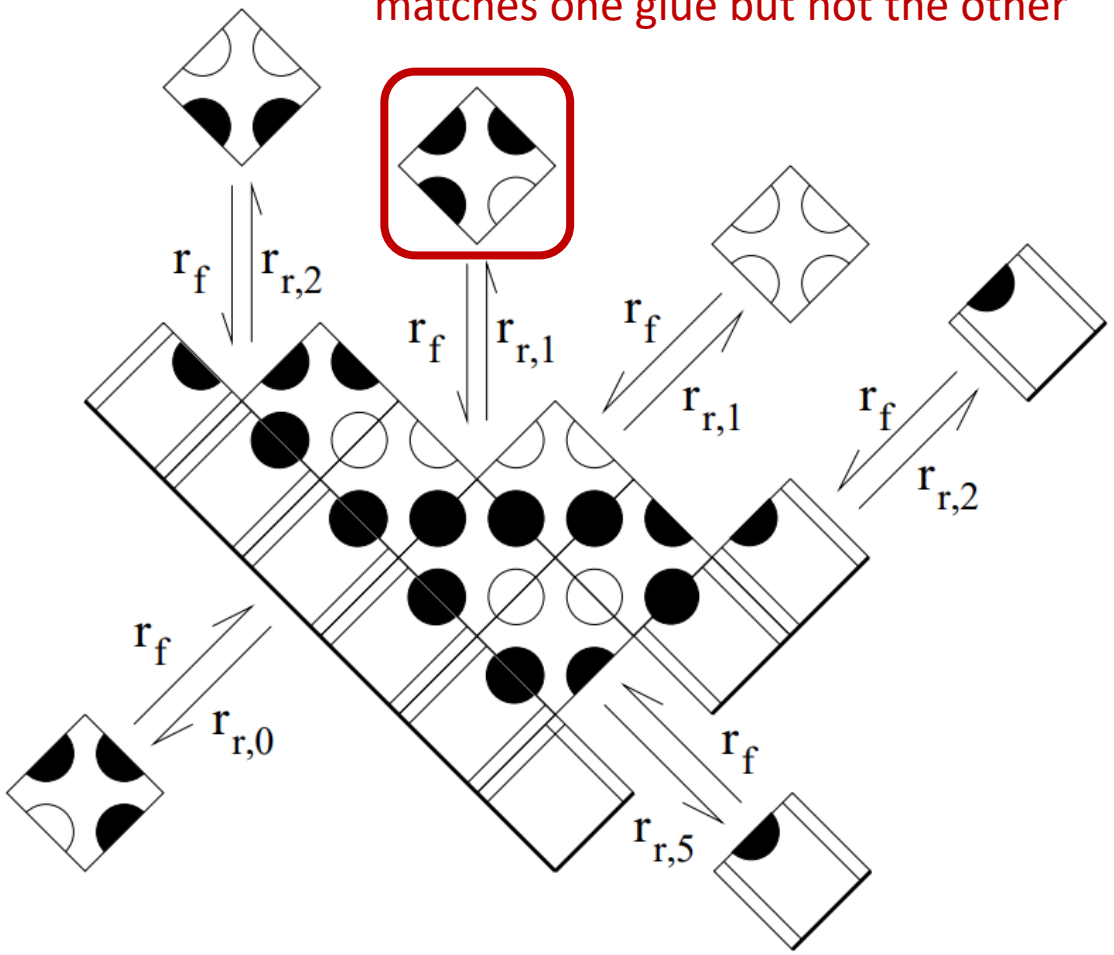

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  - a.k.a., continuous time Markov process
- Take home message: tiles bound with fewer glues (potential errors) fall off faster, but could get locked in by subsequent neighboring attachment

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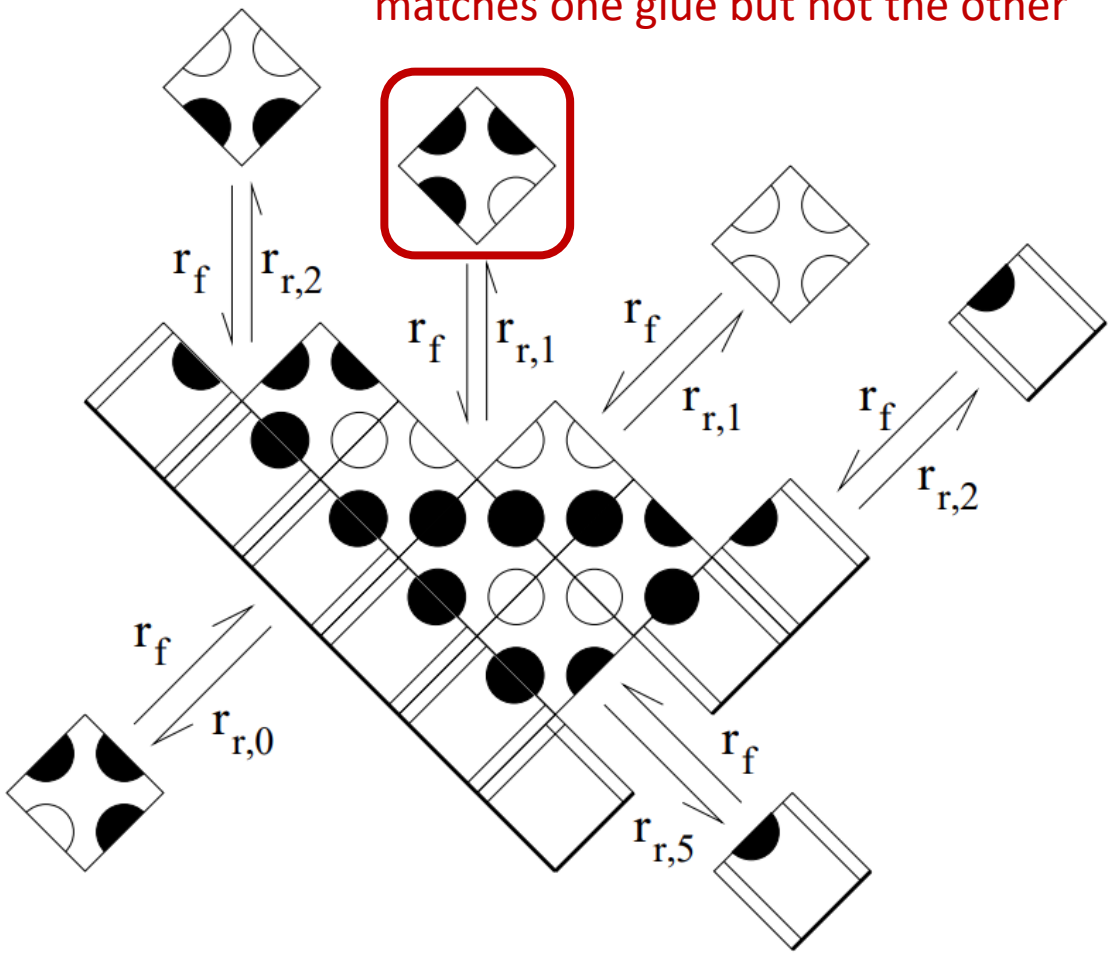

kTAM simulators

- ISU TAS (developed by Matt Patitz) also does kTAM simulation:
  - http://self-assembly.net/wiki/index.php?title=ISU_TAS
  - http://self-assembly.net/wiki/index.php?title=ISU_TAS_Tutorials
- xgrow (new version developed by Constantine Evans): https://github.com/DNA-and-Natural-Algorithms-Group/xgrow
- xgrow (original version developed by Erik Winfree)
  - https://www.dna.caltech.edu/Xgrow/
  - older and a bit less intuitive

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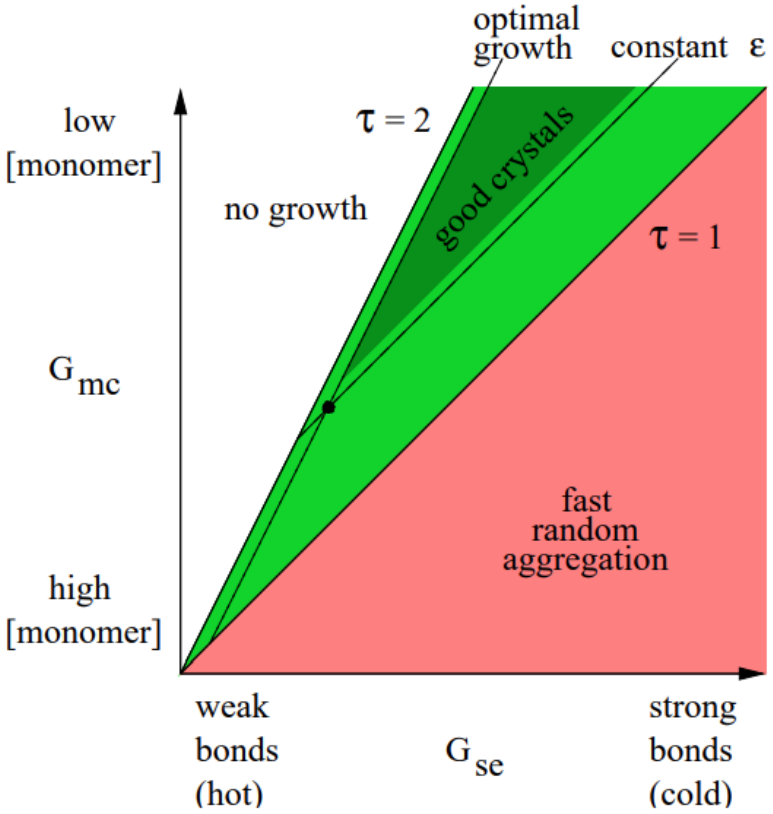

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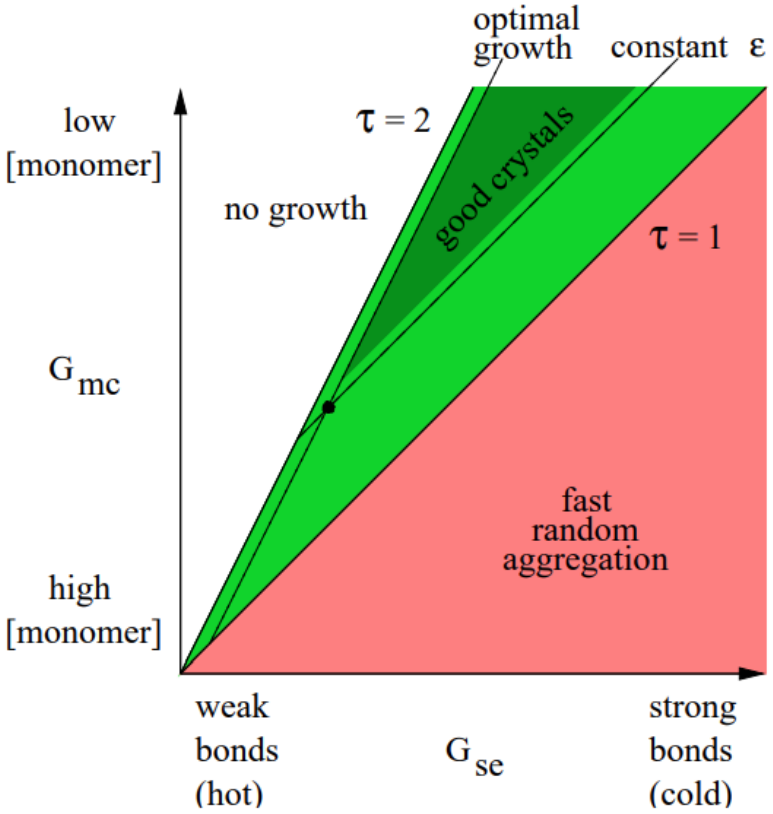

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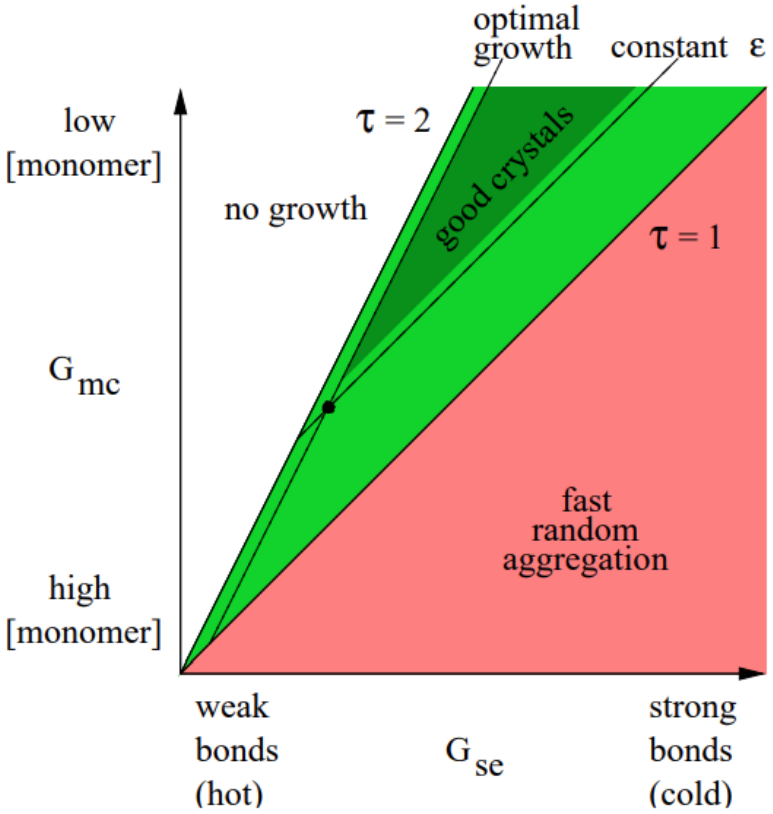

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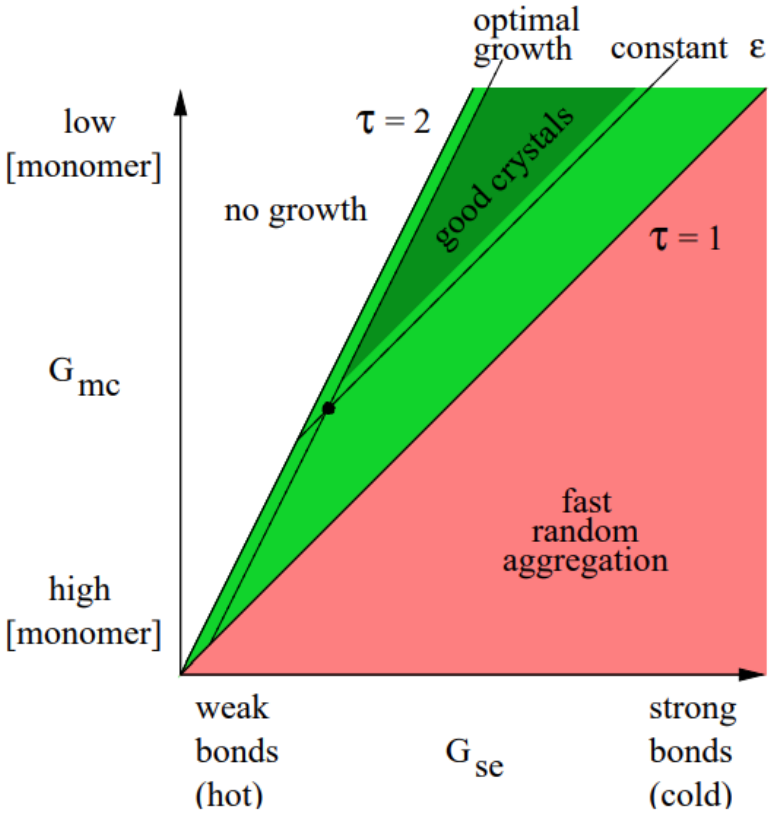
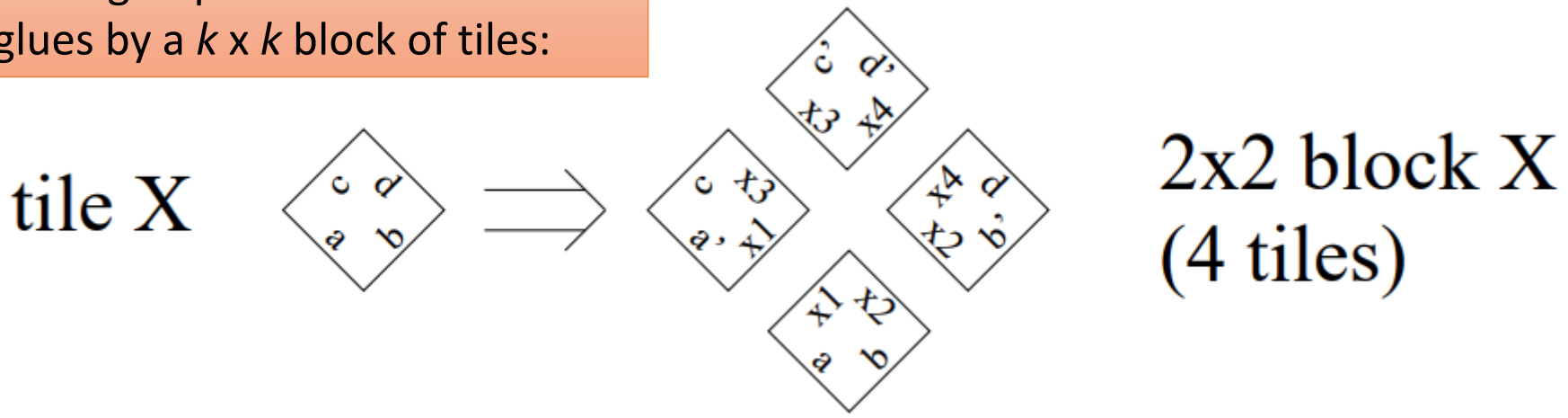
Theorem [Winfree, 1998]: To have total error rate ε, for fastest assembly speed, set $G_{se} = \ln(4/\varepsilon)$ and $G_{mc} = \ln(8/\varepsilon^2)$, i.e., $G_{mc} = 2G_{se} - \ln 2$, i.e., $r_f/r_{r,2} = 2$



Proofreading: Algorithmic error correction

k x *k* proofreading: replace each tile with all strength-1 glues by a *k* x *k* block of tiles:



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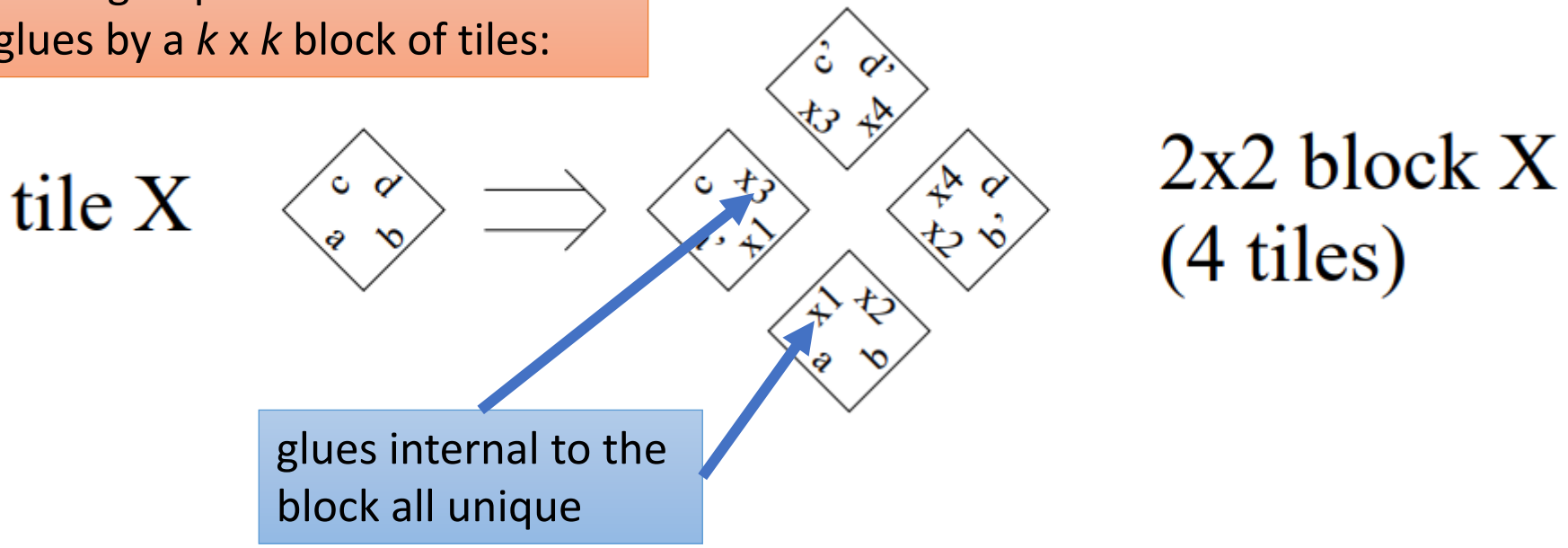

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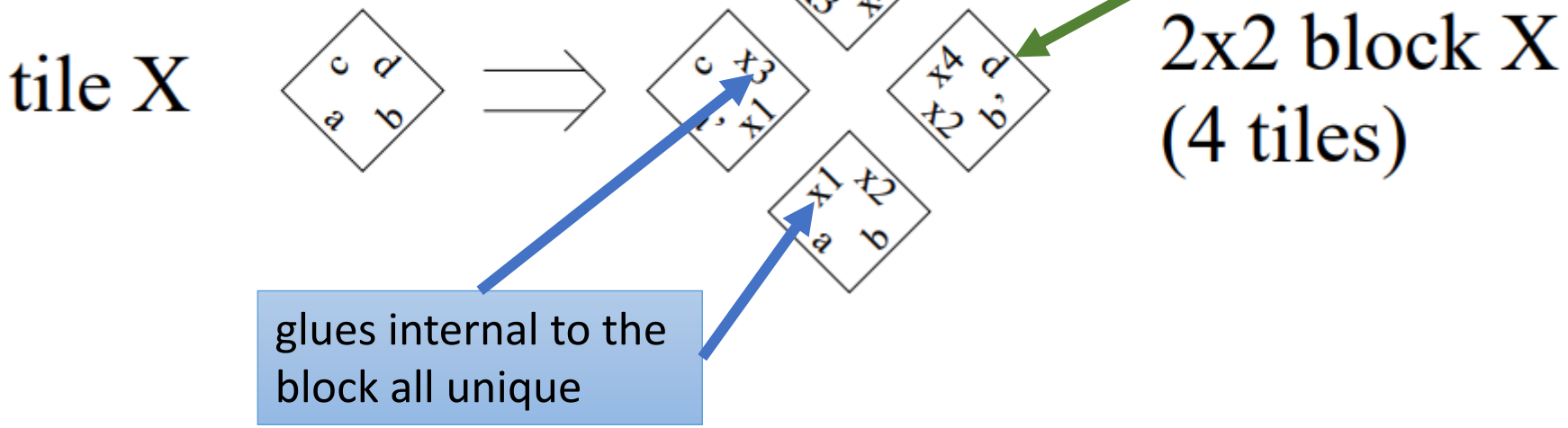

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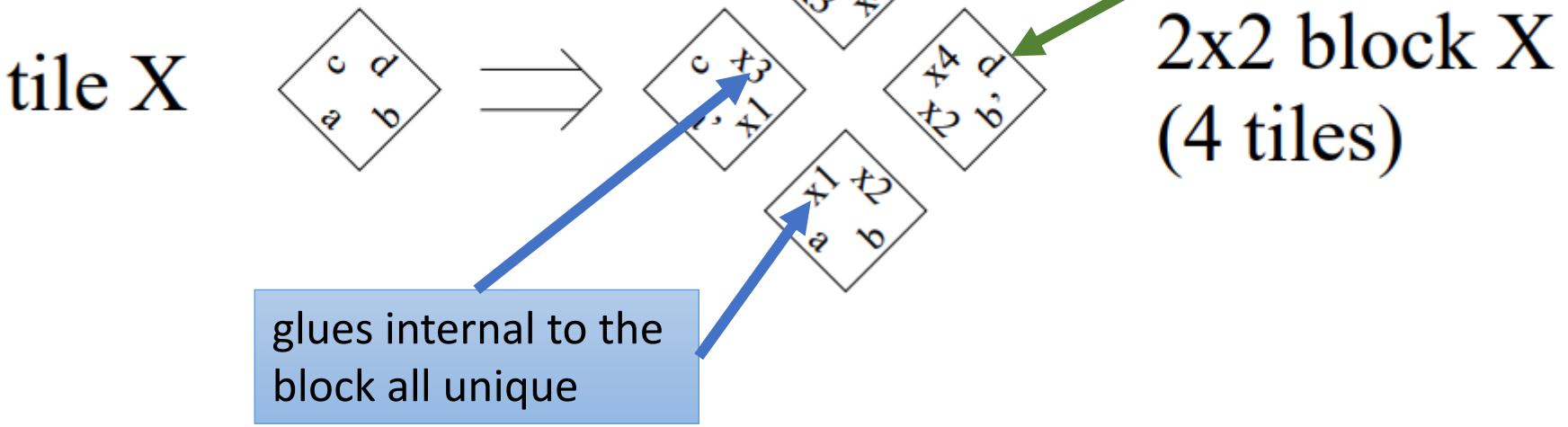

Proposition: No tiling of the *k* x *k* region with "consistent external glues" (all represent the same glue in original tile set) has *m* mismatches, where $0 < m < k$, i.e., if any mismatch occurs, then at least *k* mismatches occur before the *k* x *k* block can be completed to represent the wrong external glue.

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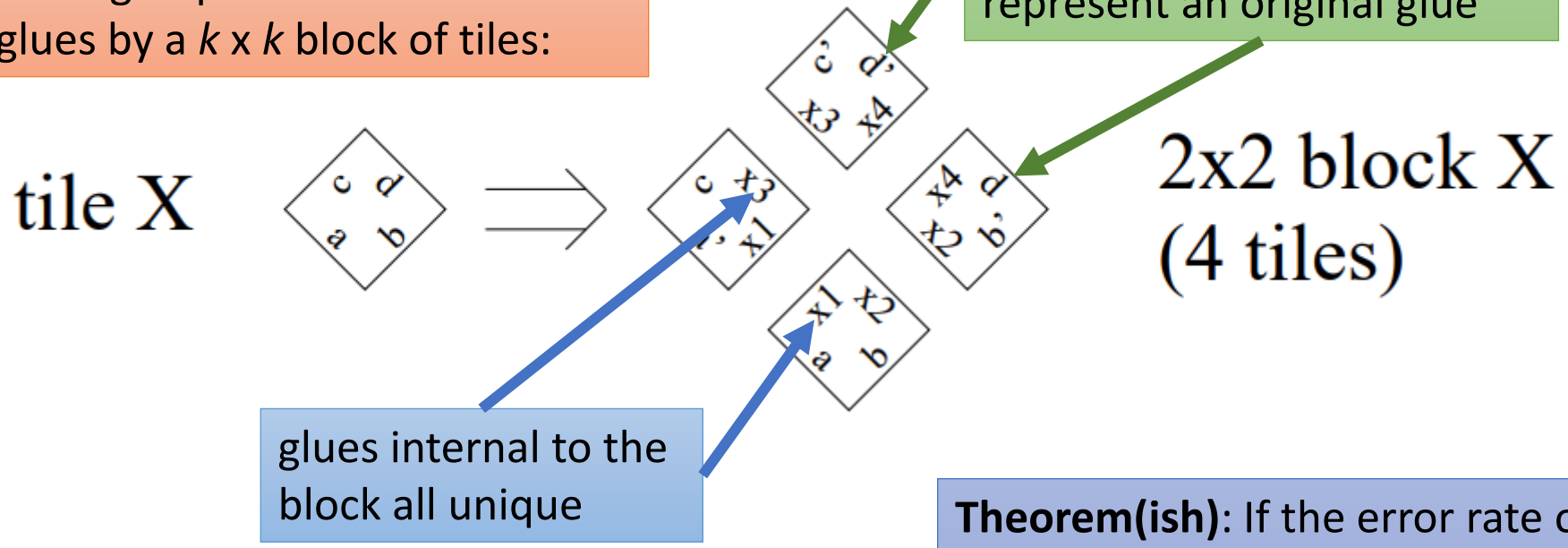

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Theorem(ish): If the error rate of the original tile system is ε, the error rate of the *k* x *k* proofreading tile system is $O(\varepsilon^k)$, e.g., if $\varepsilon = 0.01$, then 2 x 2 proofreading gets error rate about $\varepsilon^2 = 0.0001$.

Experimental algorithmic self-assembly

Crystals that think
about how they're growing

joint work with Damien Woods, Erik Winfree, Cameron Myhrvold, Joy Hui, Felix Zhou, Peng Yin

slides for ECS 232: Theory of Molecular Computation

Caltech Inria Paris UC Davis Harvard

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Caltech

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Harvard

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(co-first author)

Erik Winfree

co-authors

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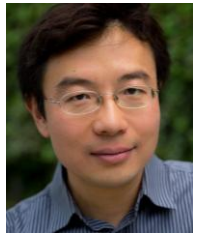
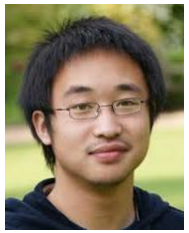


Felix Zhou



Joy Hui

lab/science help

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Yannick Rondolez
Mingjie Dai
Nikhil Gopalkrishnan
Chris Thachuk
Nadine Dabby
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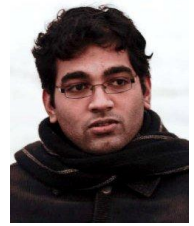
Paul Rothemund
Bryan Wei
Cody Geary
Ashwin Gopinath



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CRA Computing Research Association

CCC Computing Community Consortium

Diverse and robust molecular algorithms using reprogrammable DNA self-assembly.
Damien Woods†, David Doty†, Cameron Myhrvold, Joy Hui, Felix Zhou, Peng Yin, Erik Winfree.
Nature 2019. †*These authors contributed equally*.

Hierarchy of abstractions

➡ **Bits:** **Boolean circuits compute**

Tiles: Tile growth implements circuits

DNA: DNA strands implement tiles

Harmonious arrangement

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Harmonious arrangement

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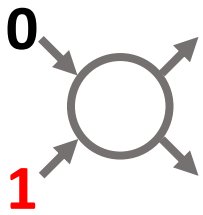

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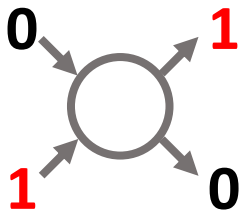

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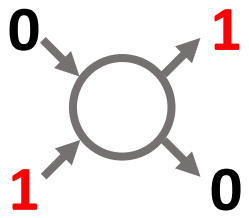

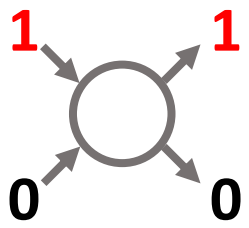

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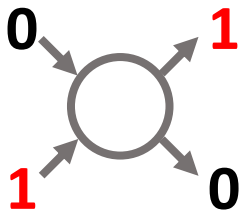

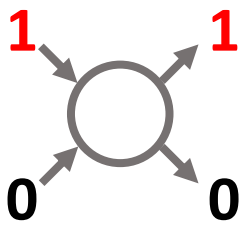

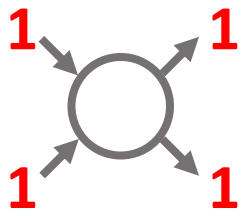

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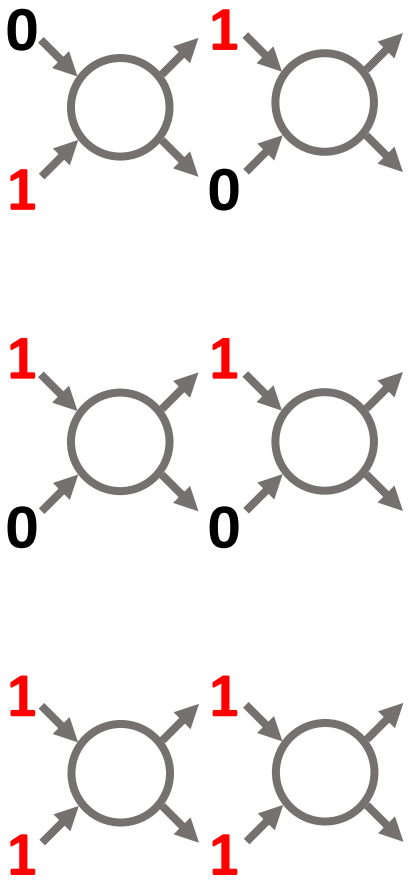

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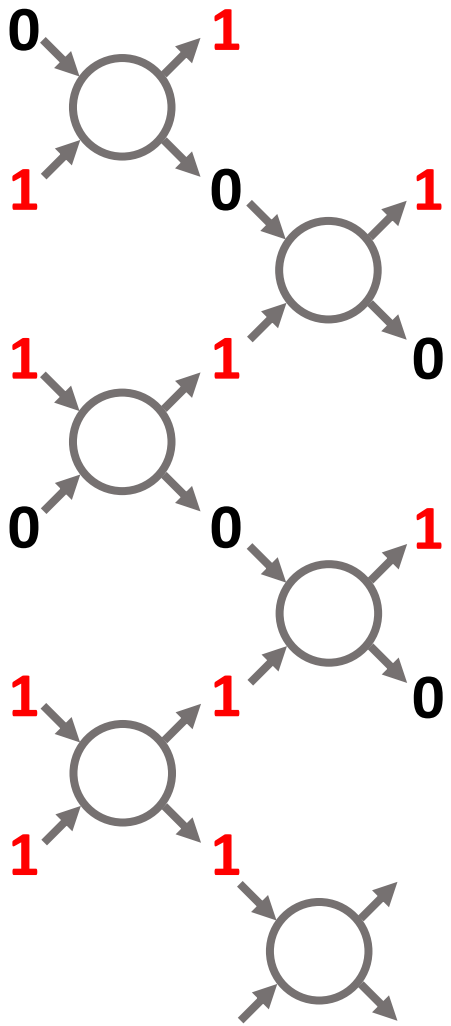

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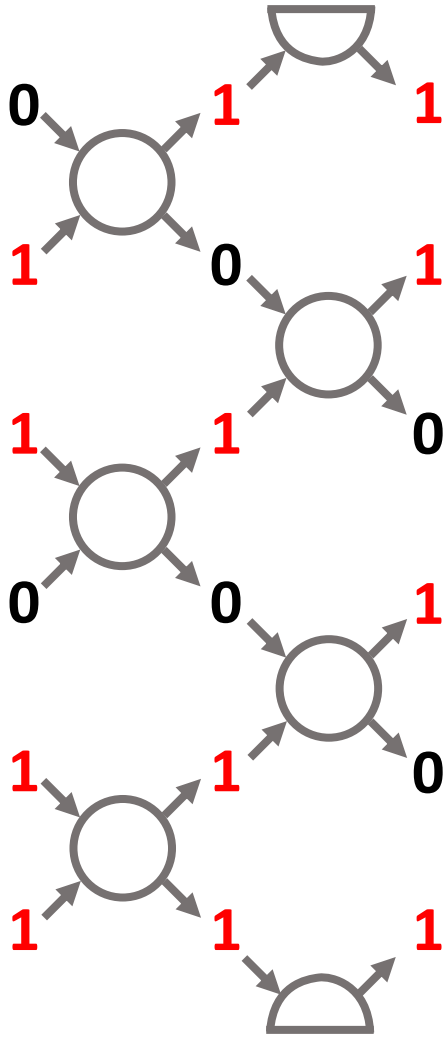

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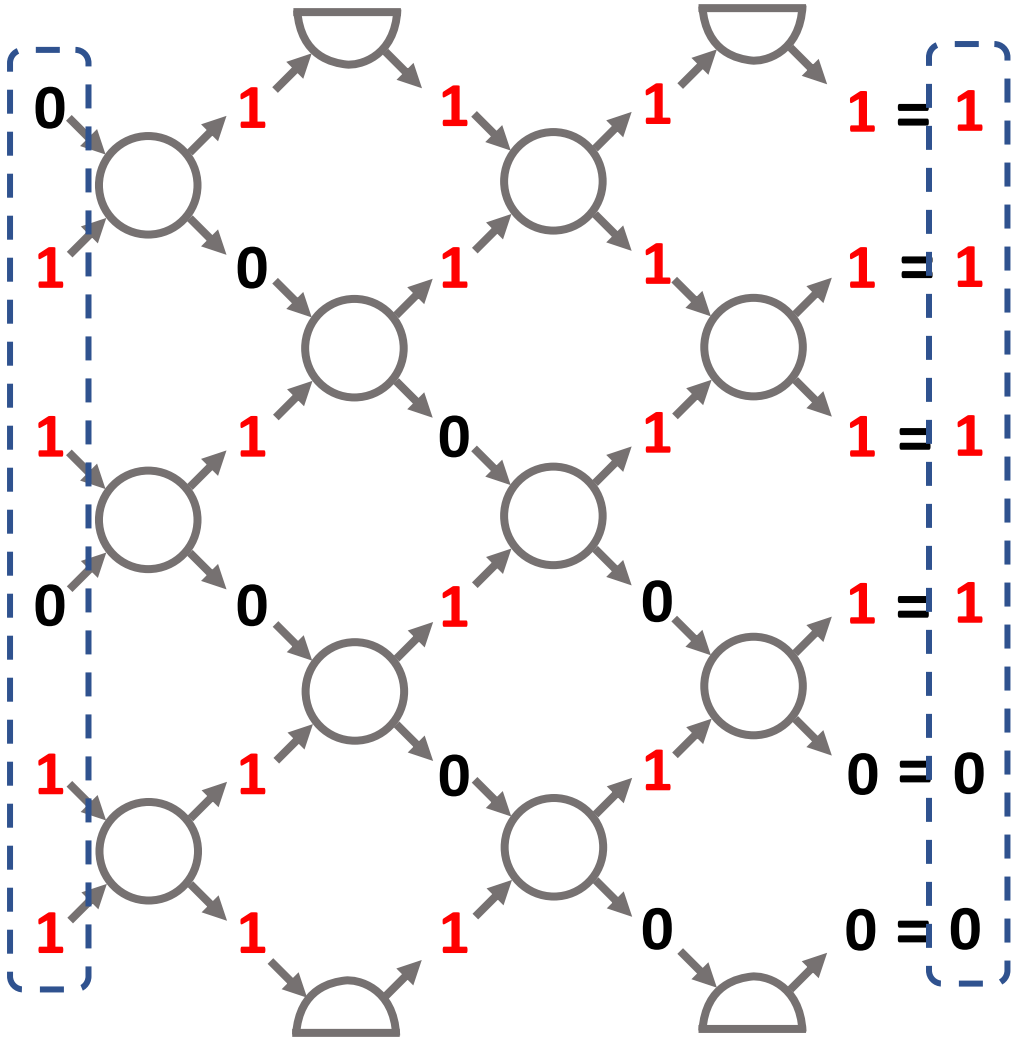
0

Harmonious arrangement



a.k.a. sorting

Odd bits

1

0

1

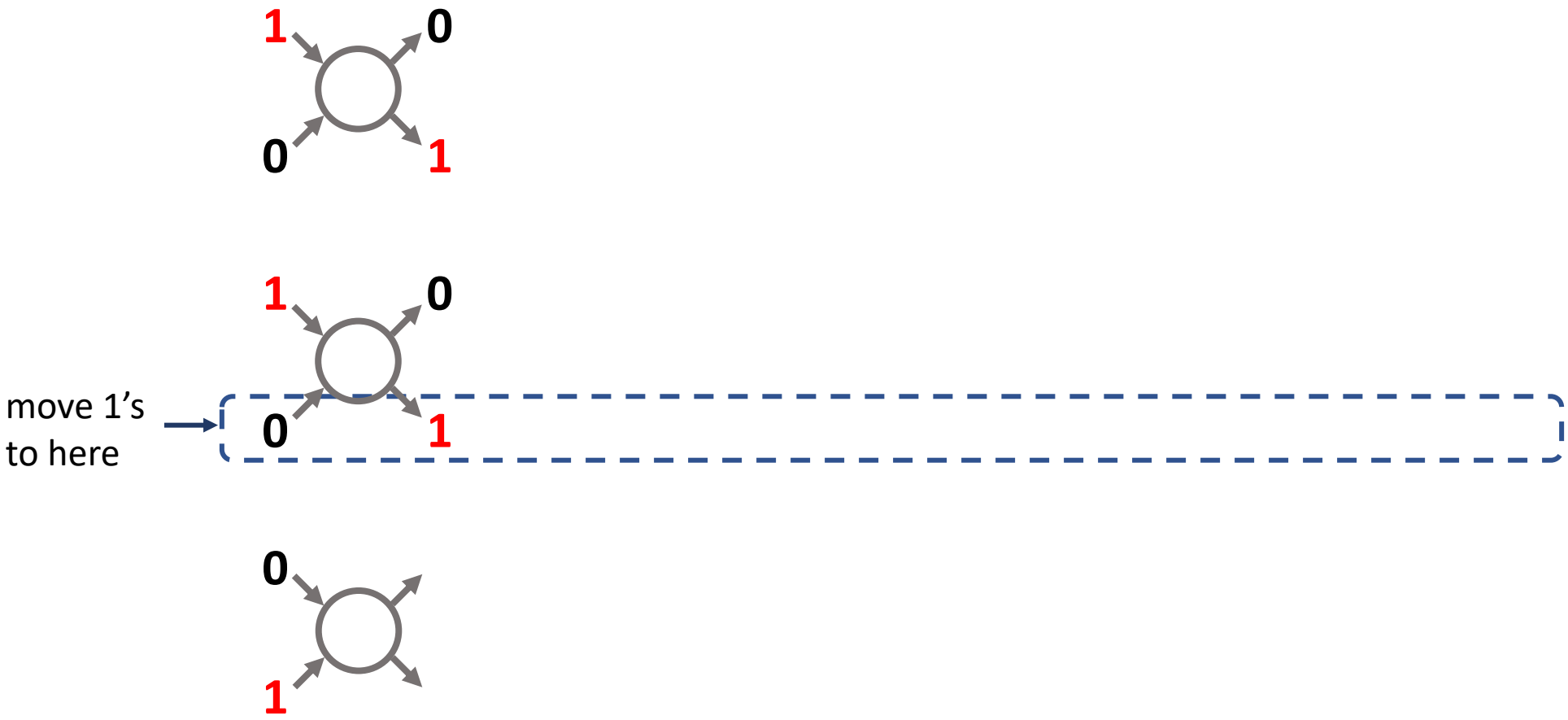
0

0

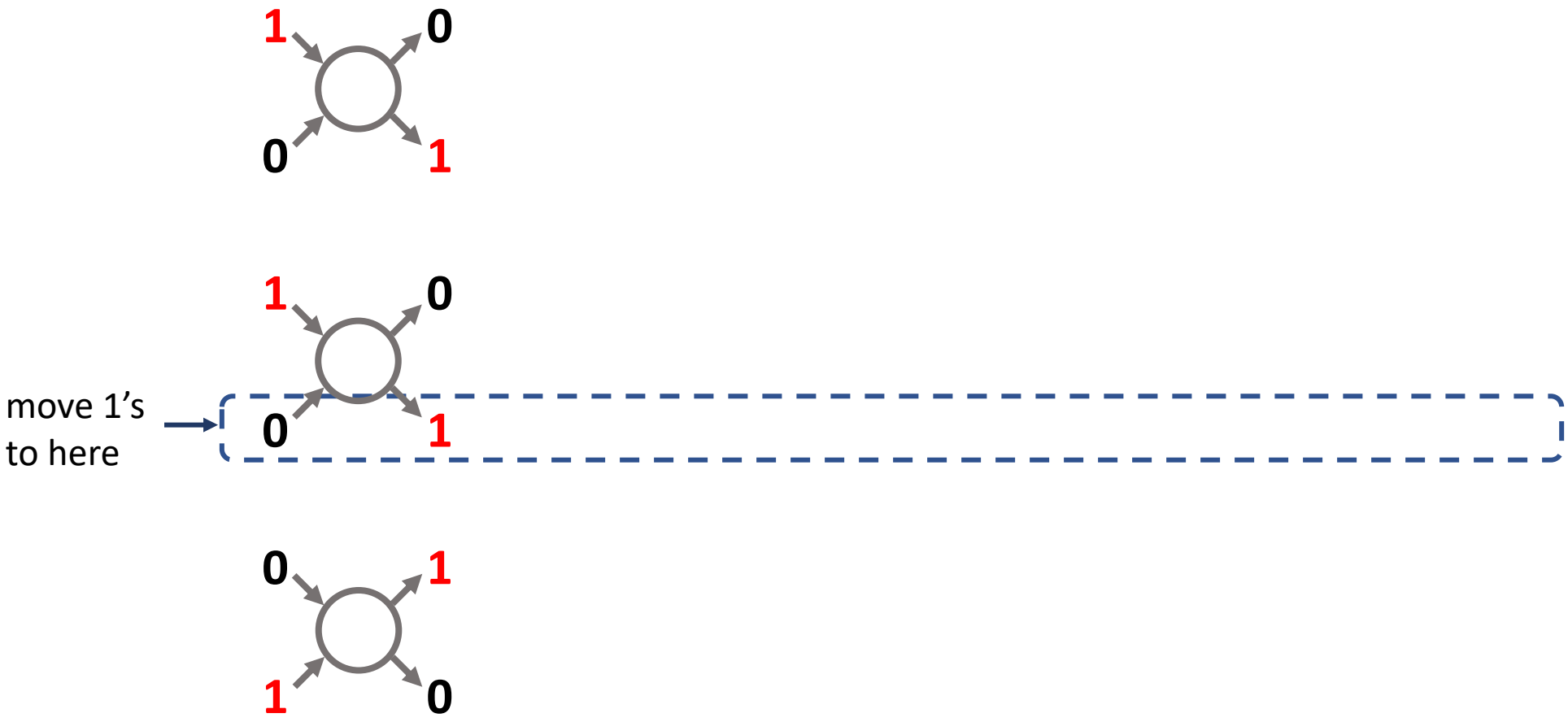
1

Odd bits

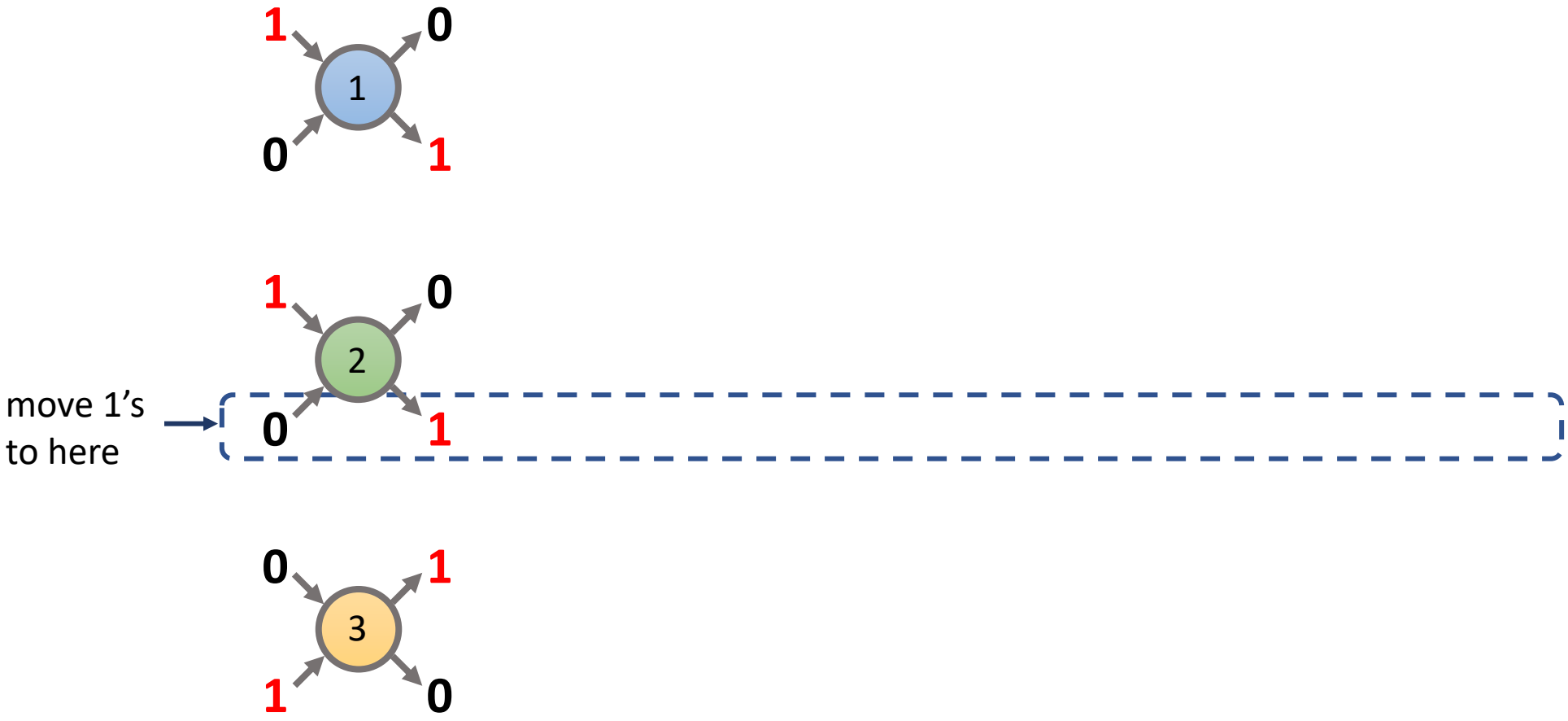
Odd bits



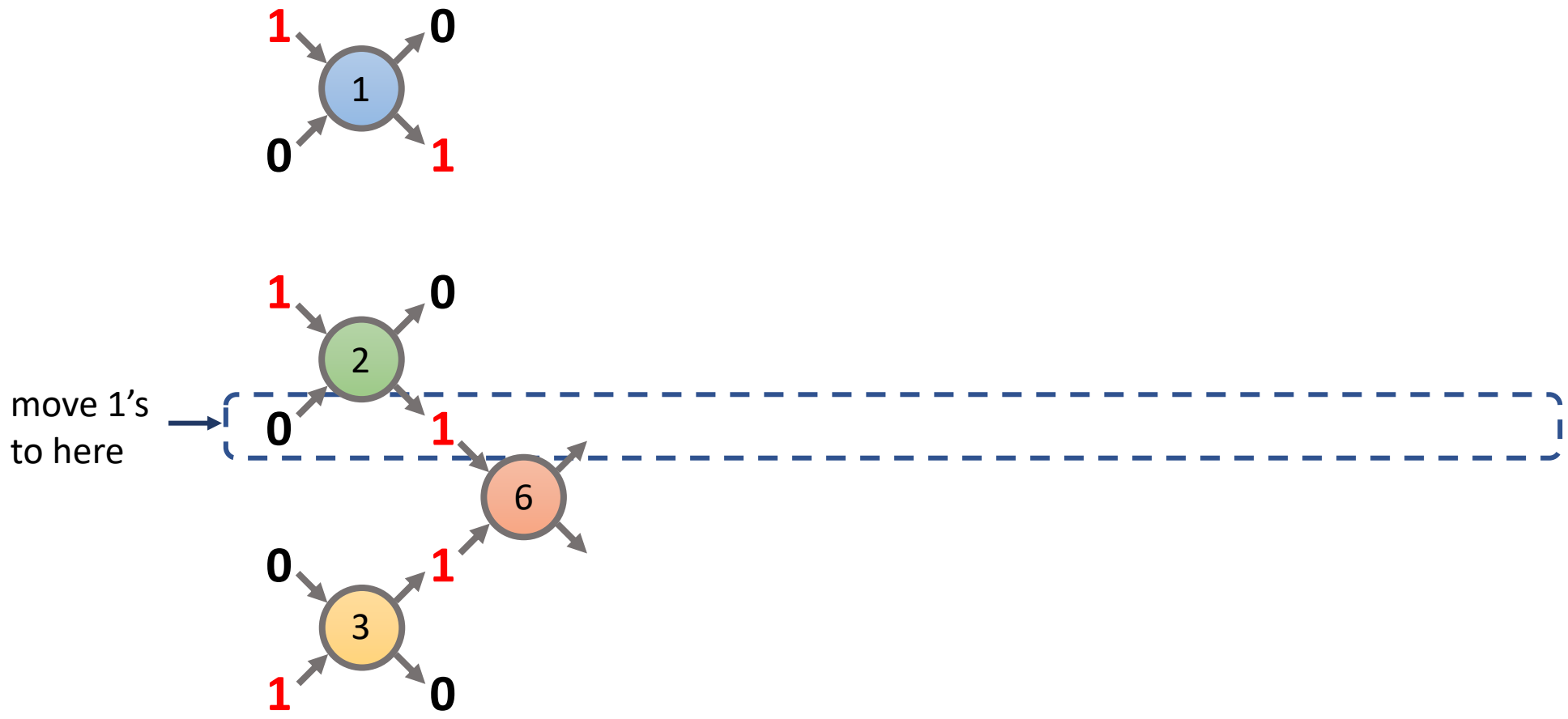
Odd bits



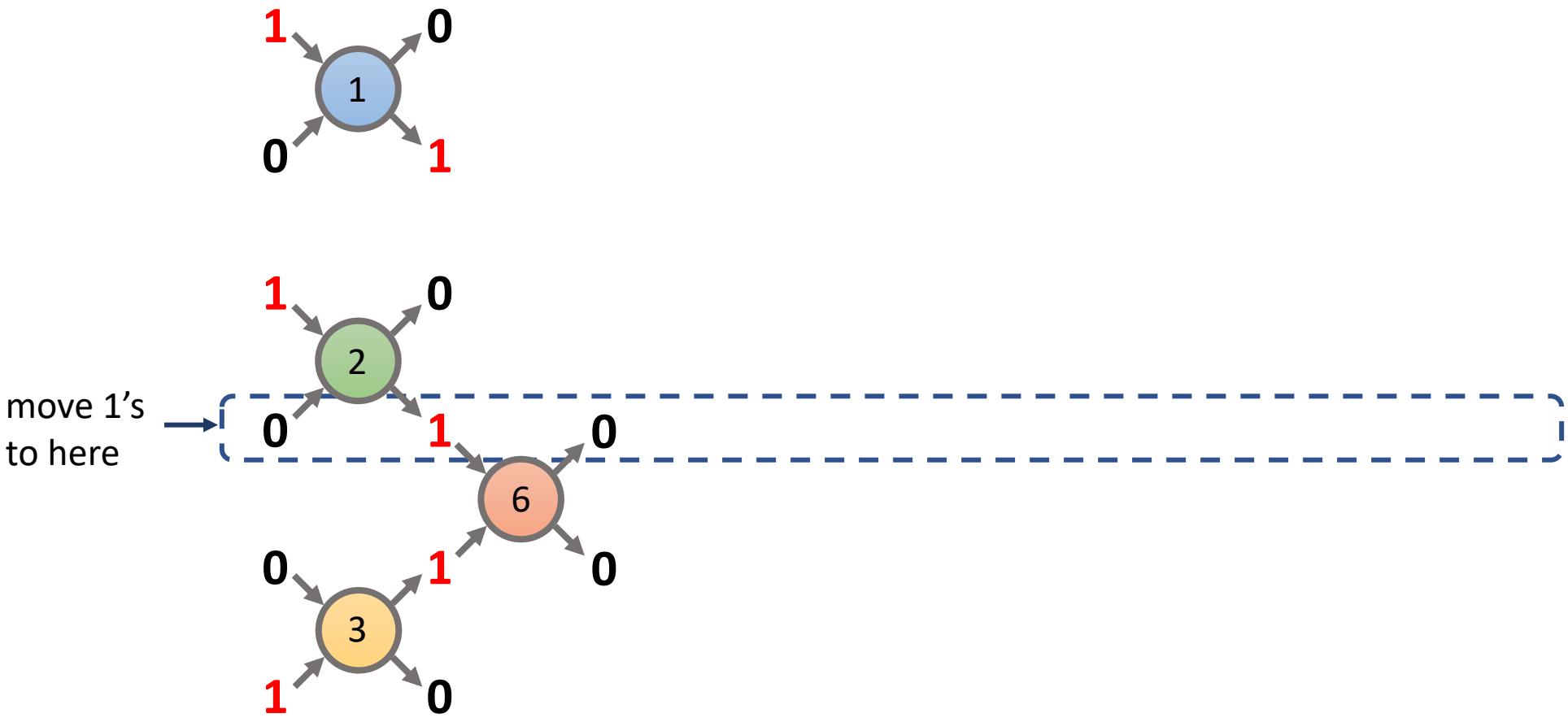
Odd bits



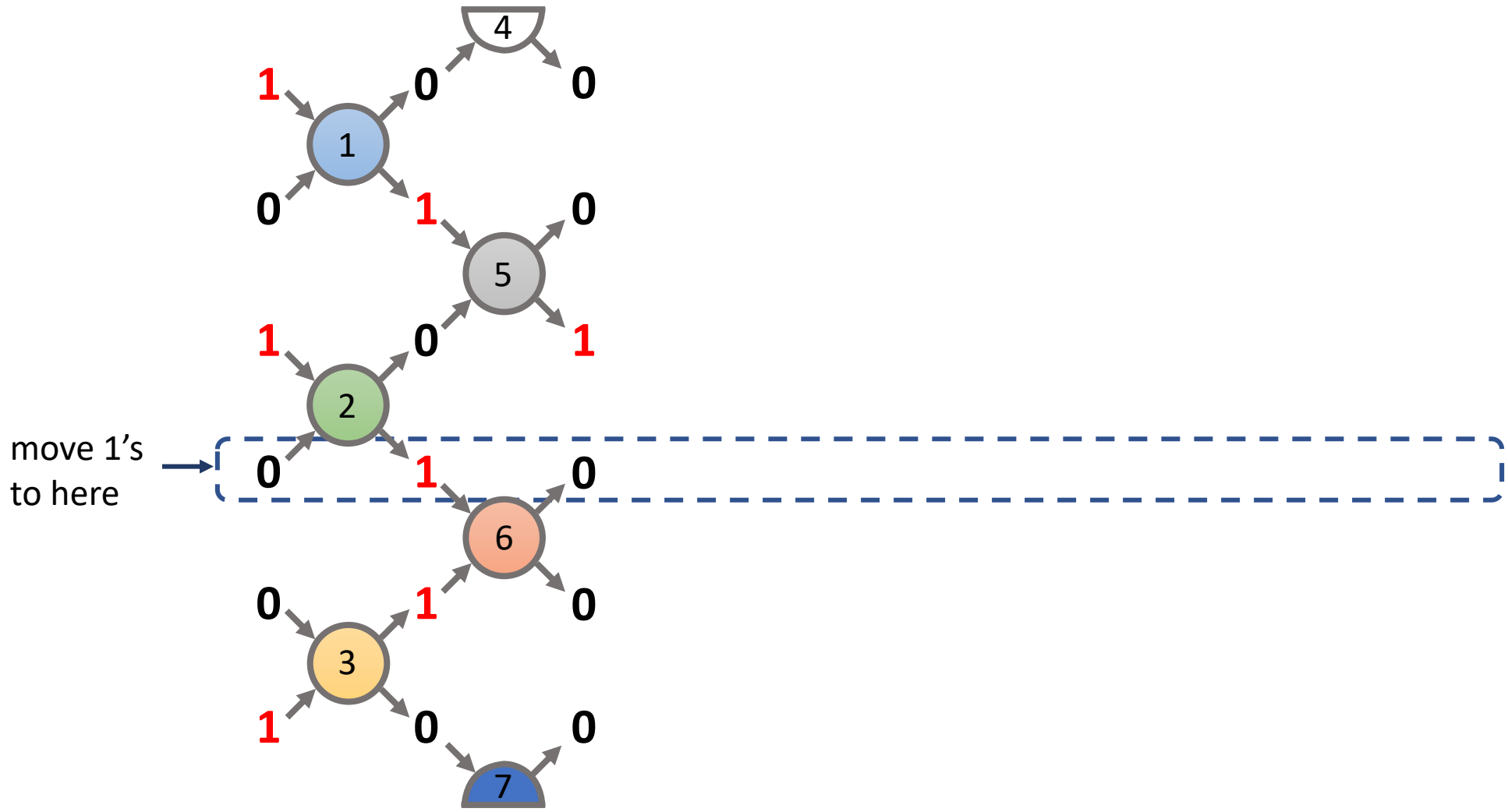
Odd bits



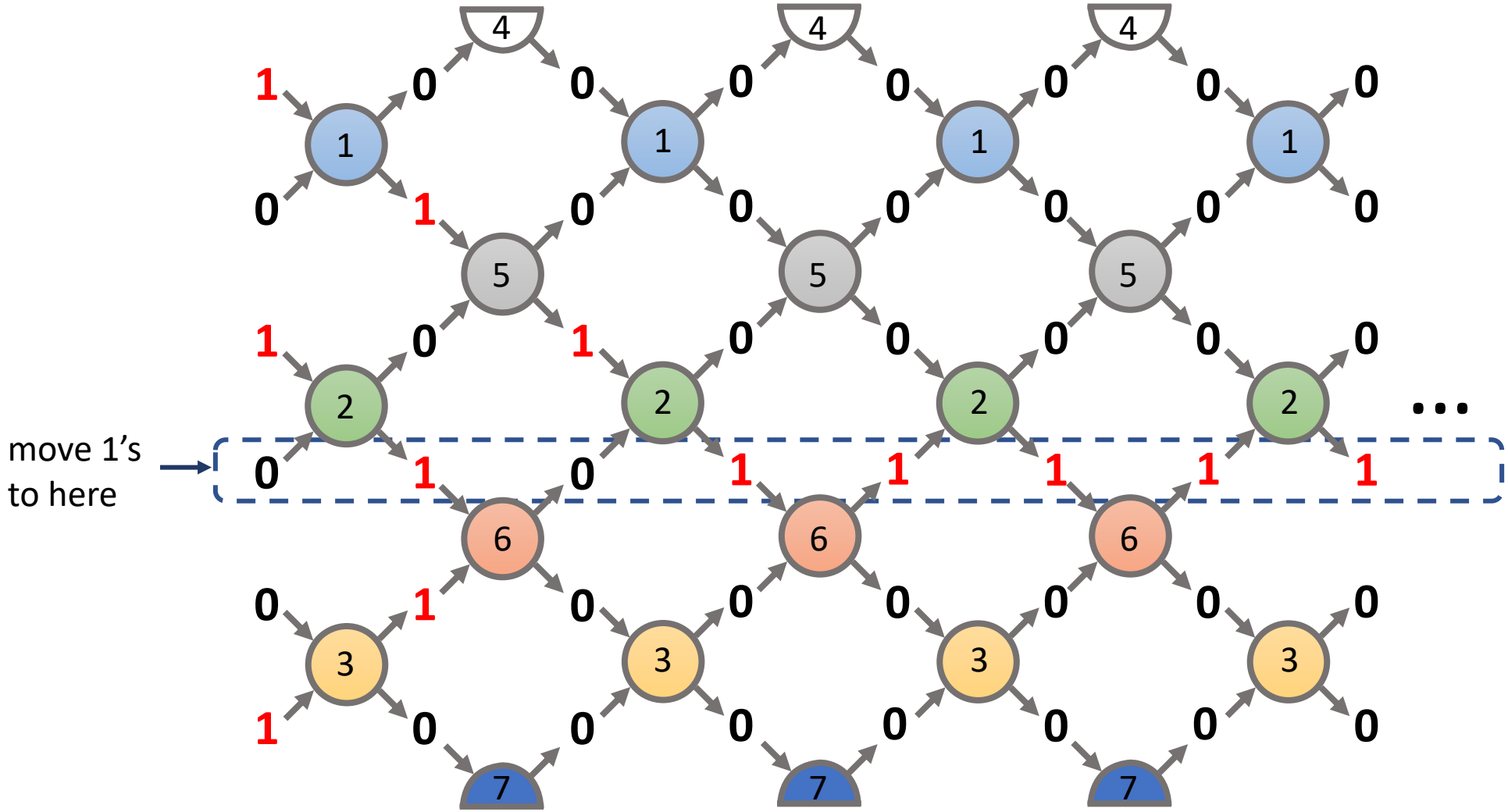
Odd bits



Odd bits

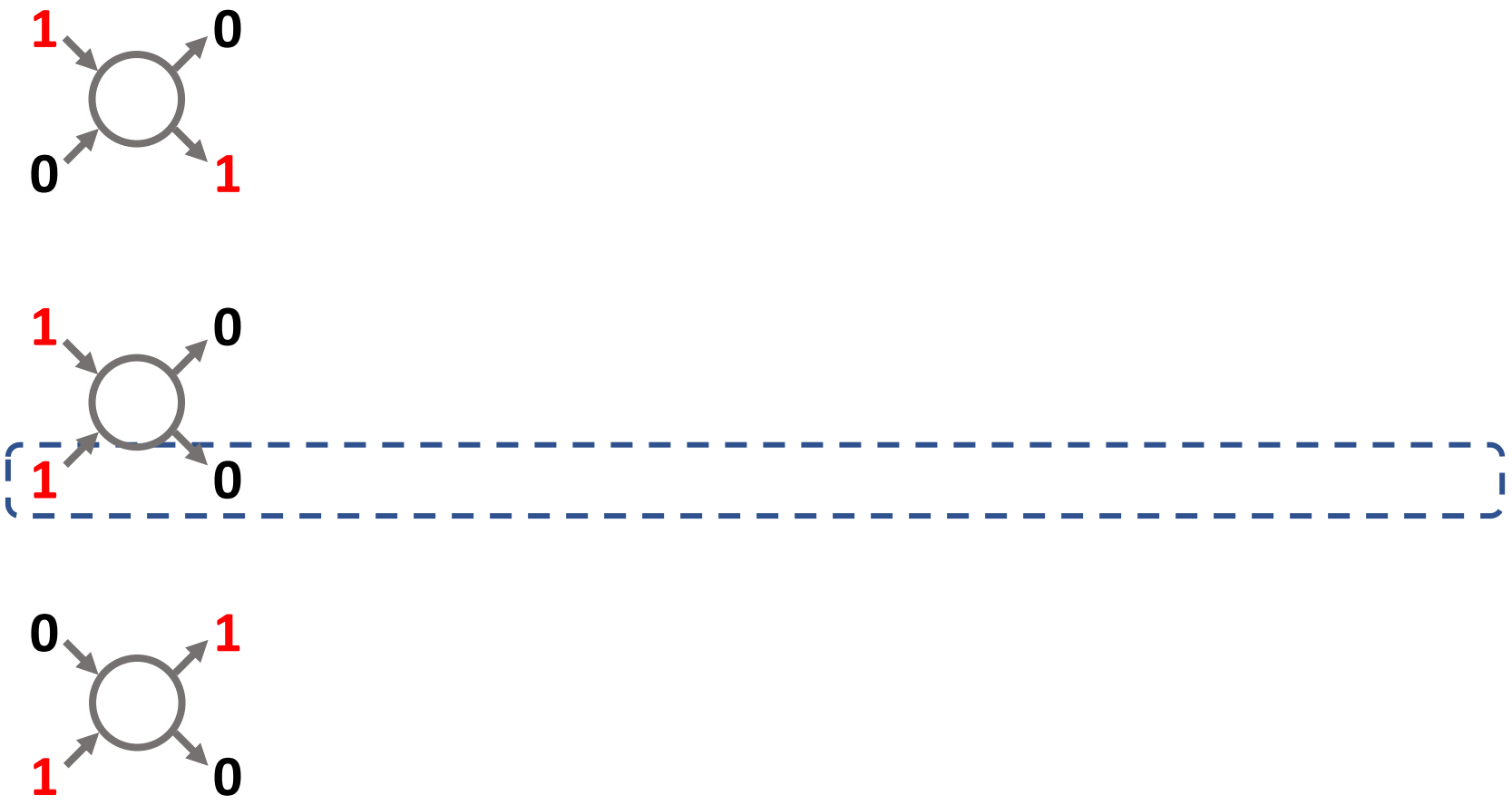


Odd bits a.k.a. parity

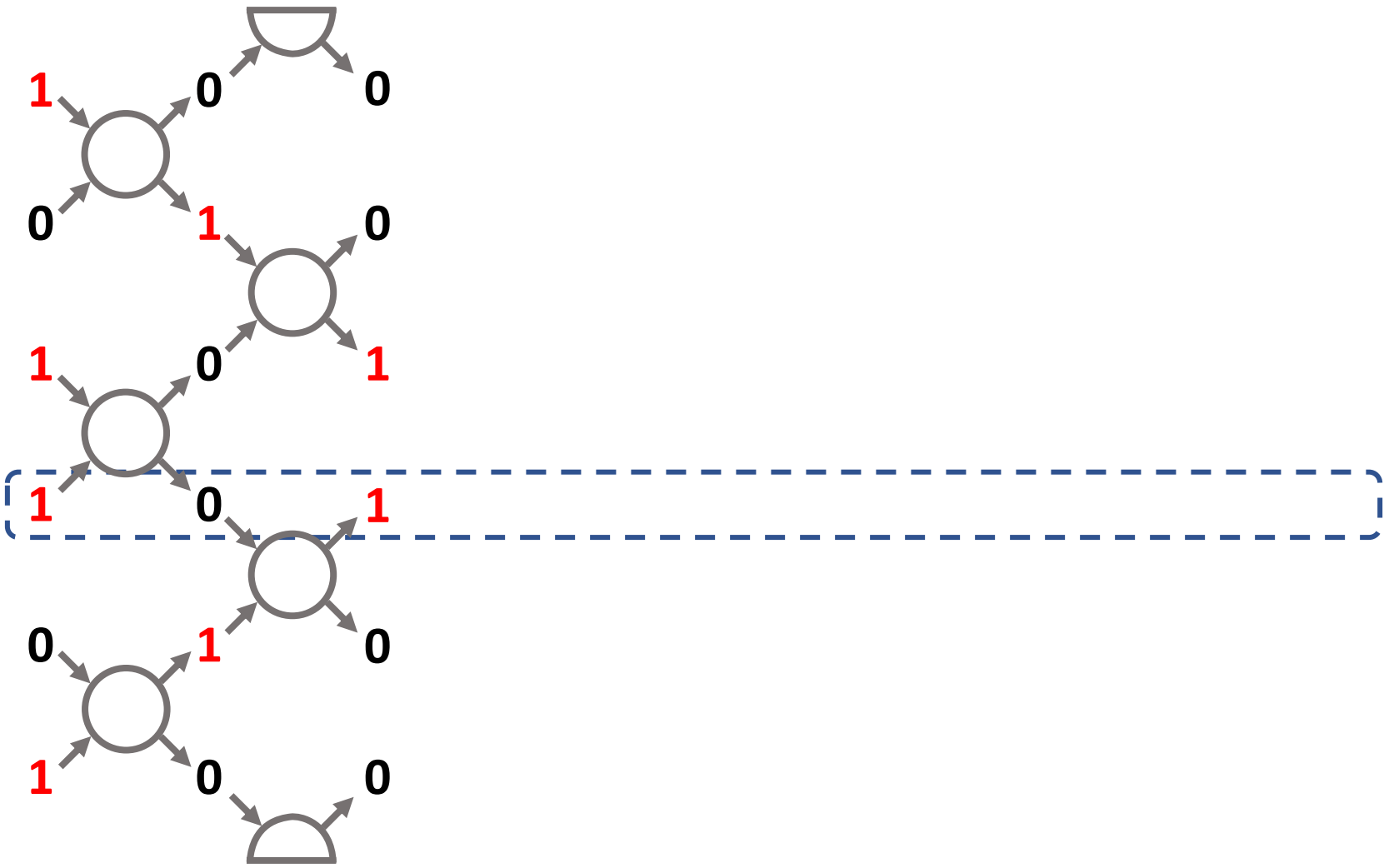


Parity

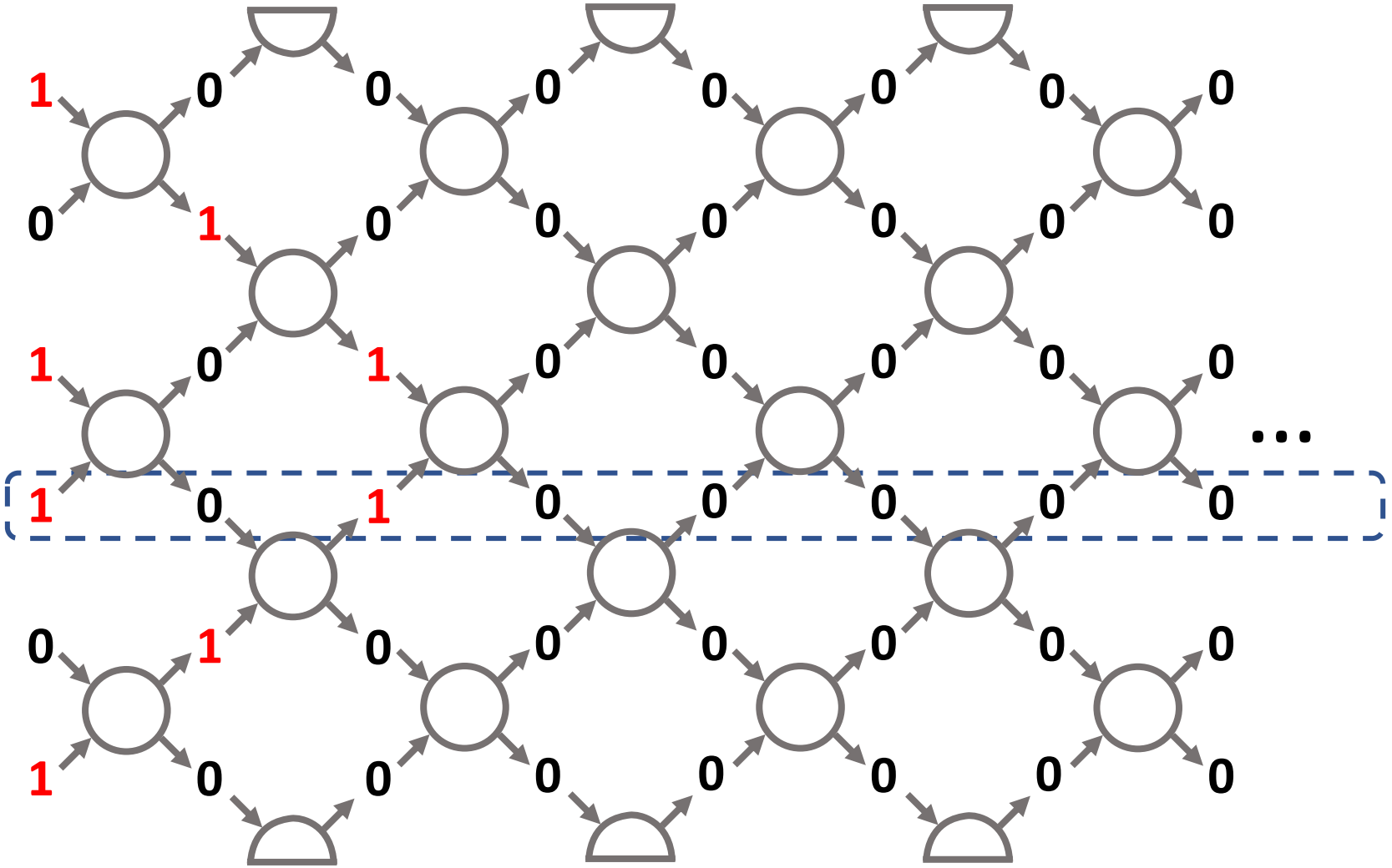
Parity



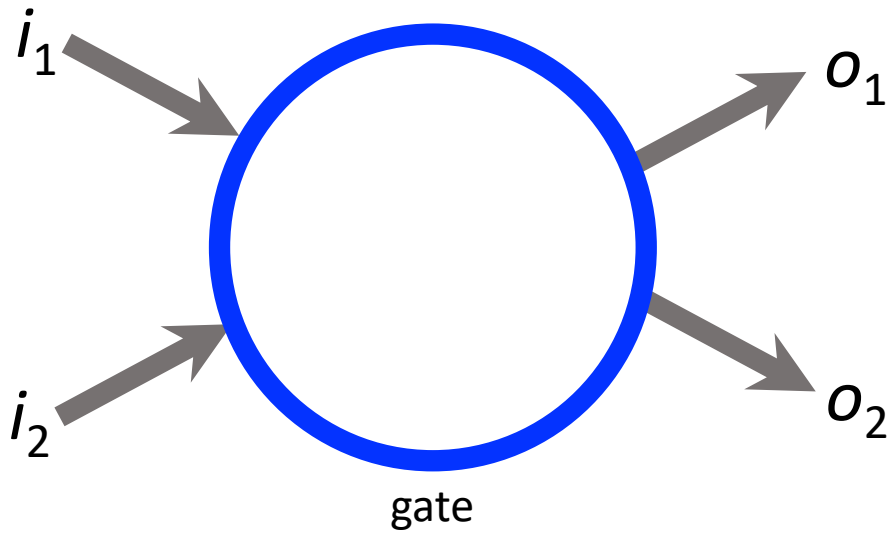
Parity



Parity

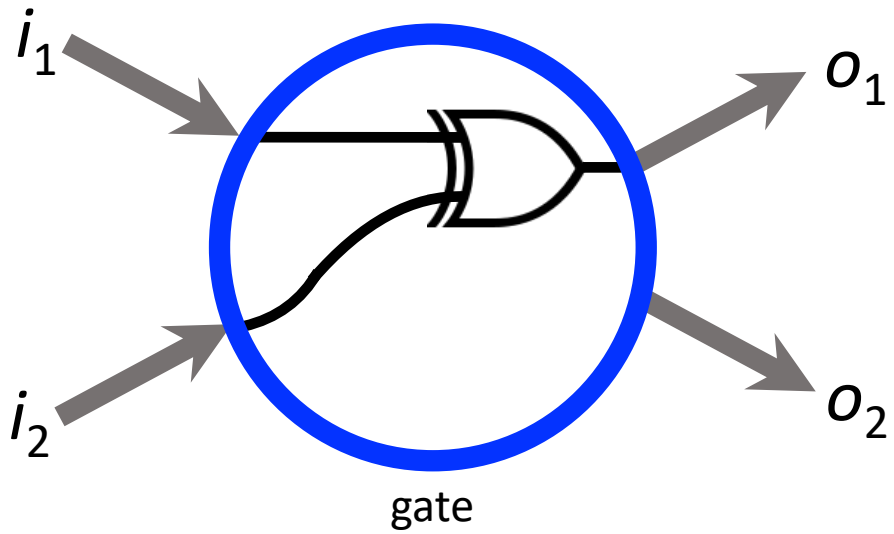


Circuit model



gate: function with two input bits i_1,i_2
and two output bits o_1,o_2

Circuit model

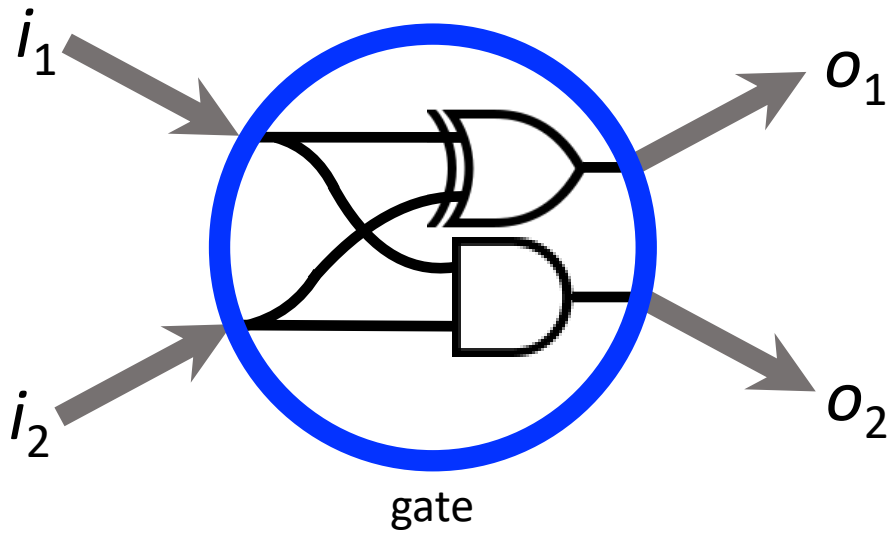


truth table

i_1	i_2	o_1	o_2
0	0	0	
0	1	1	
1	0	1	
1	1	0	

gate: function with two input bits i_1,i_2
and two output bits o_1,o_2

Circuit model

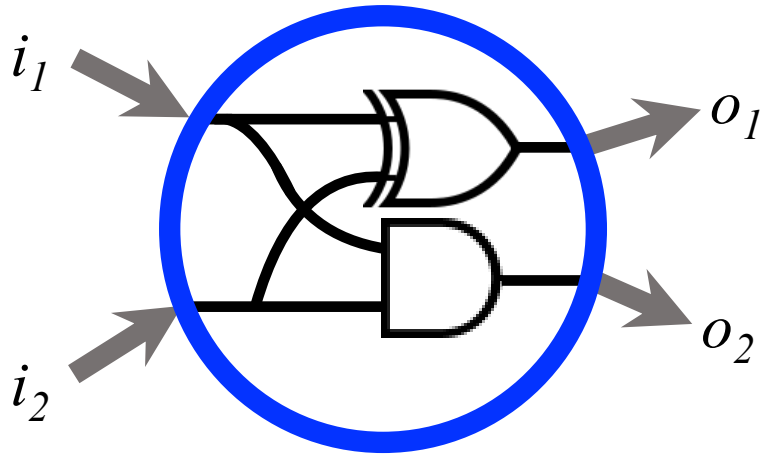


truth table

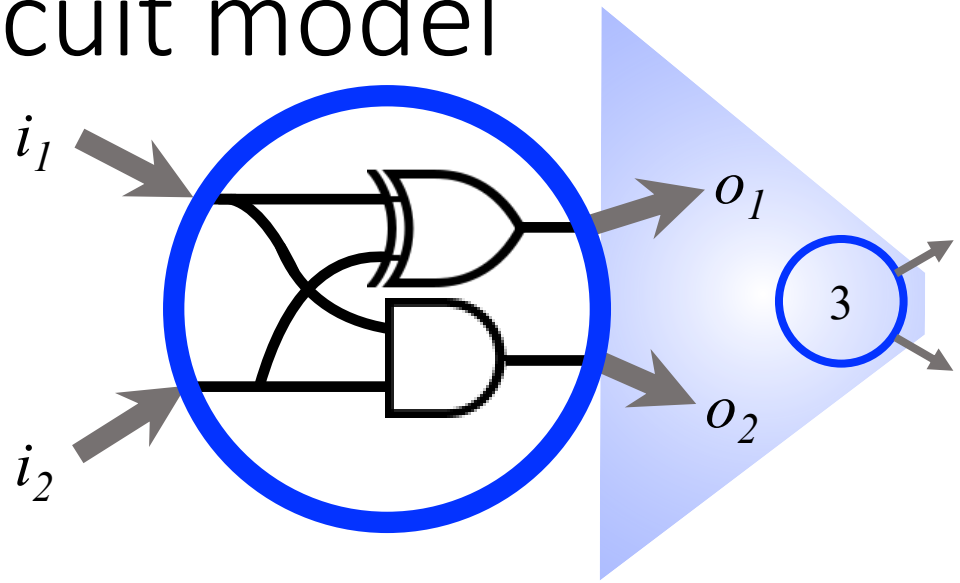
i_1	i_2	o_1	o_2
0	0	0	0
0	1	1	0
1	0	1	0
1	1	0	1

gate: function with two input bits i_1,i_2
and two output bits o_1,o_2

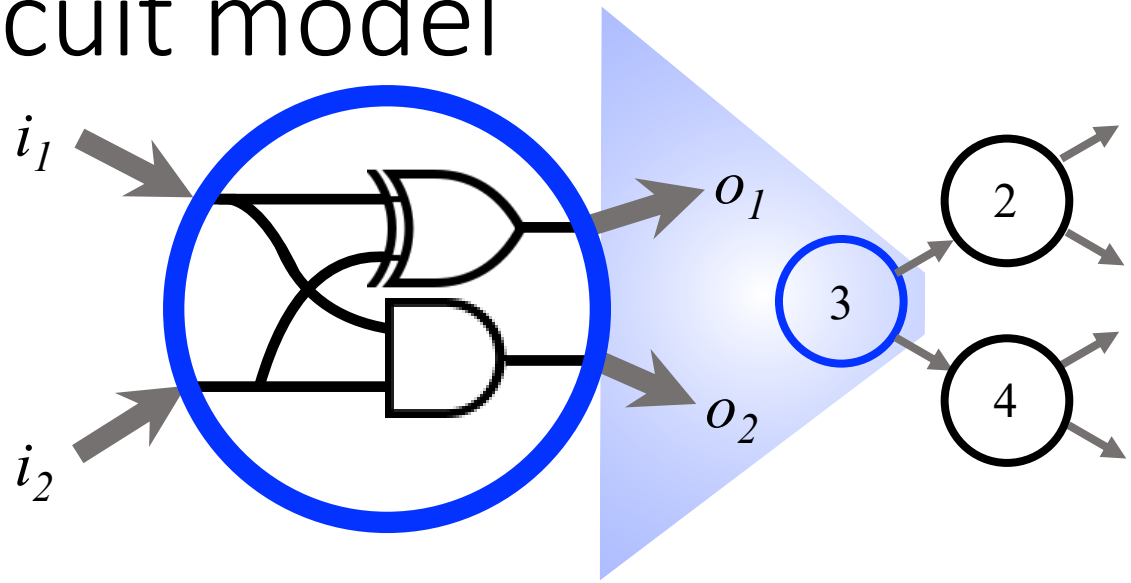
Circuit model



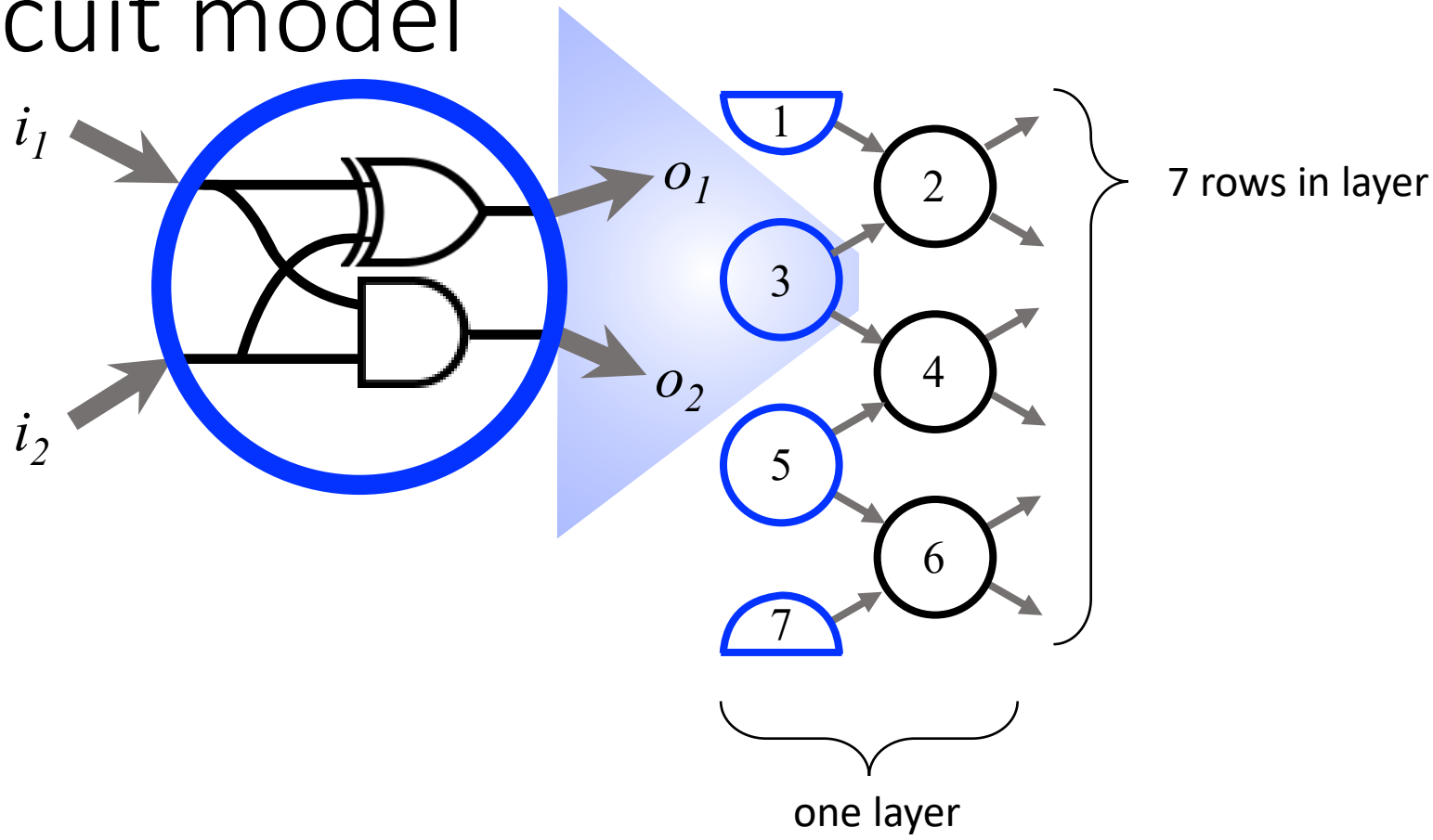
Circuit model



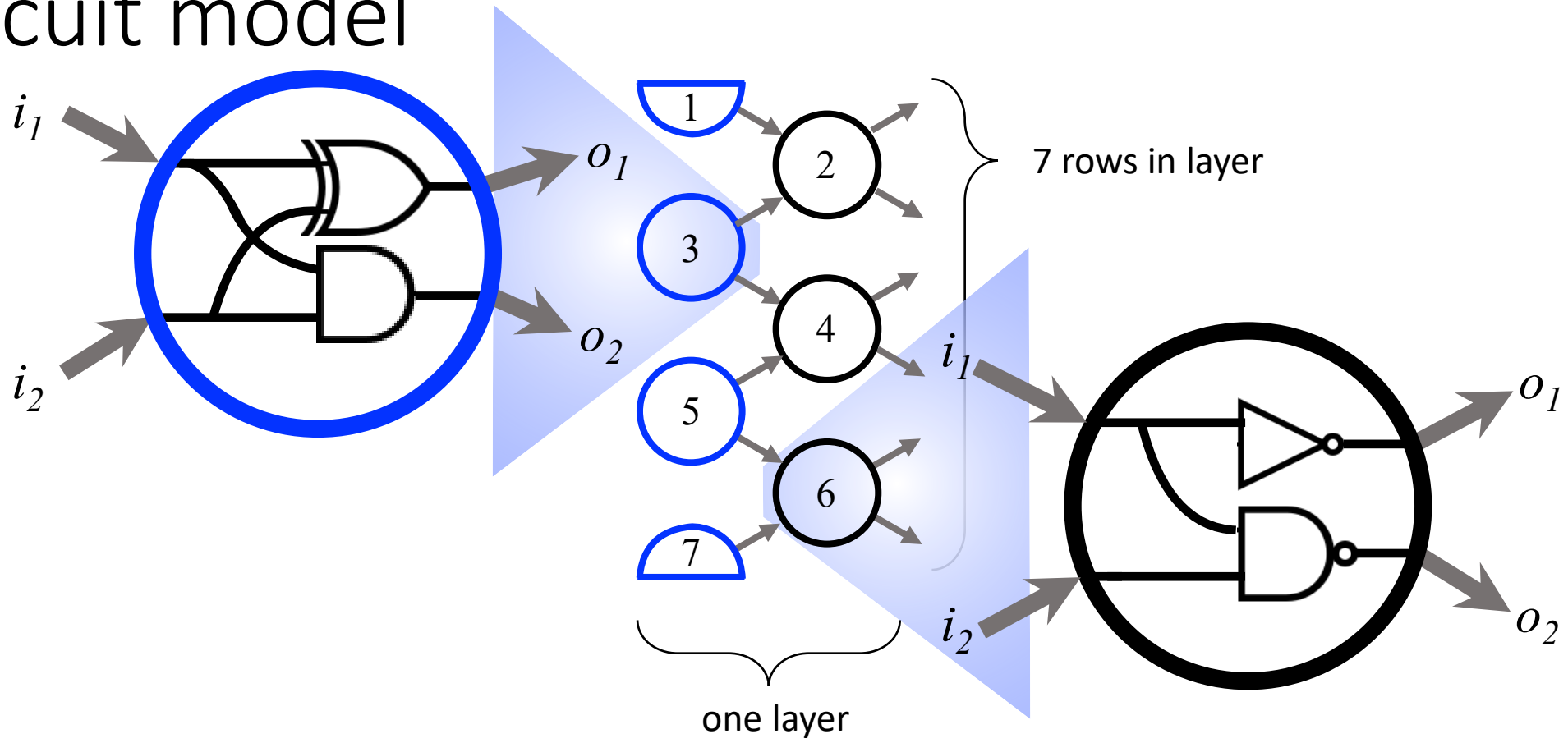
Circuit model



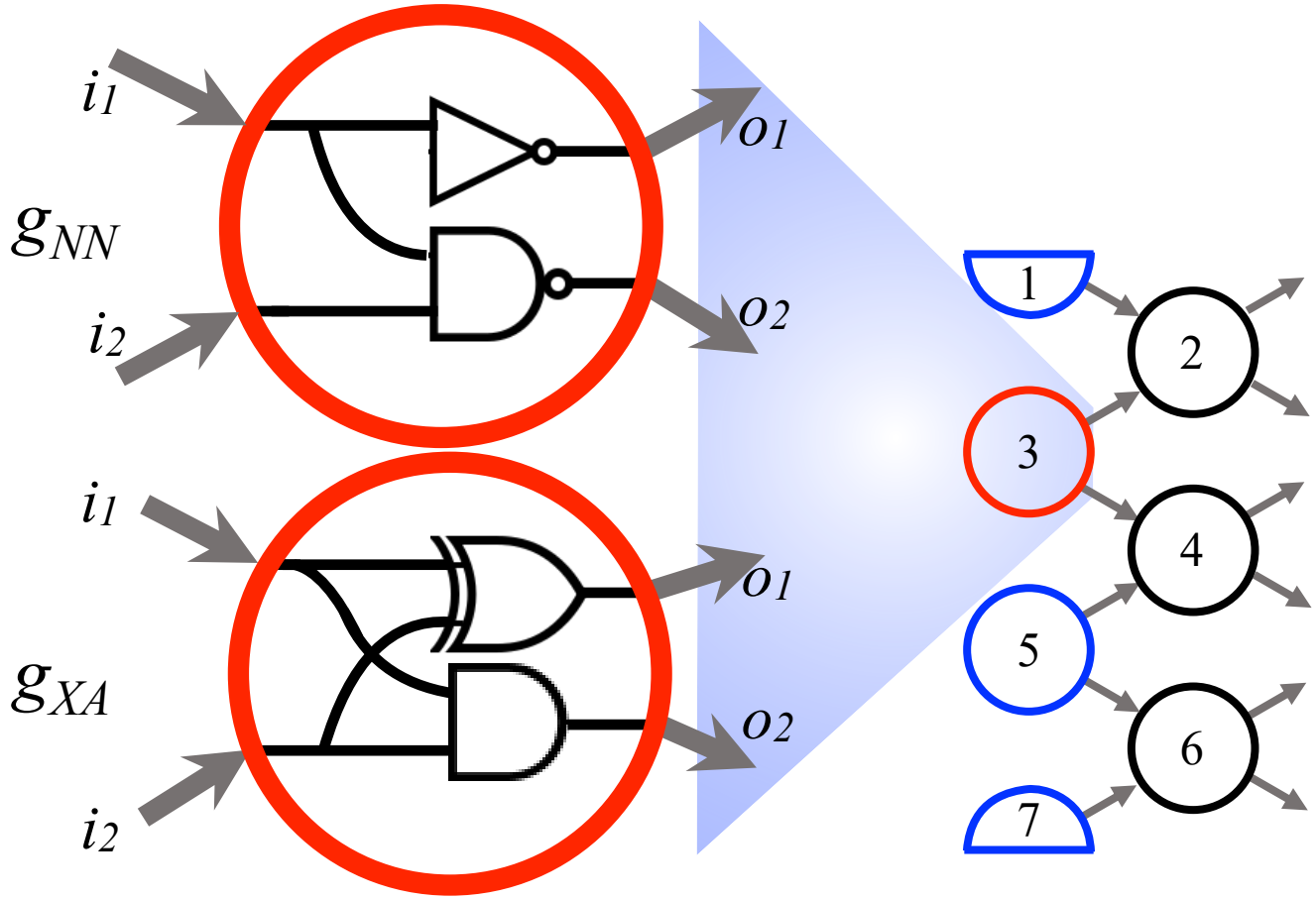
Circuit model



Circuit model



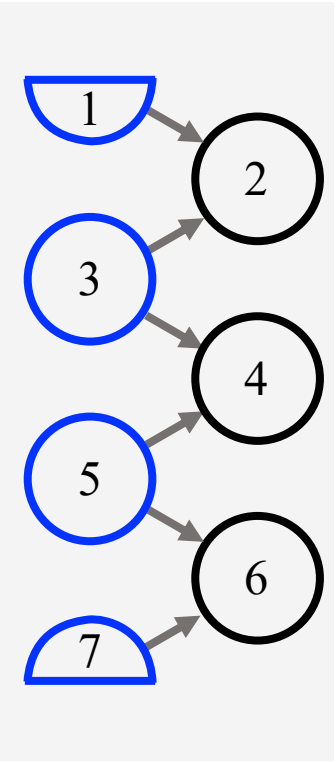
Circuit model



Randomization: Each row may be assigned ≥ 2 gates, with associated probabilities, e.g., $\Pr[g_{NN}] = \Pr[g_{XA}] = ½$

Circuit model

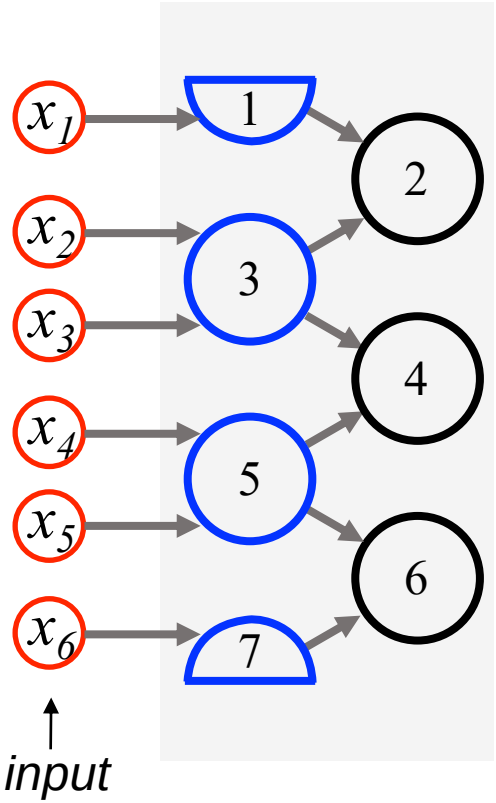
Programmer specifies layer:
gates to go in each row



Circuit model

Programmer specifies layer: gates to go in each row

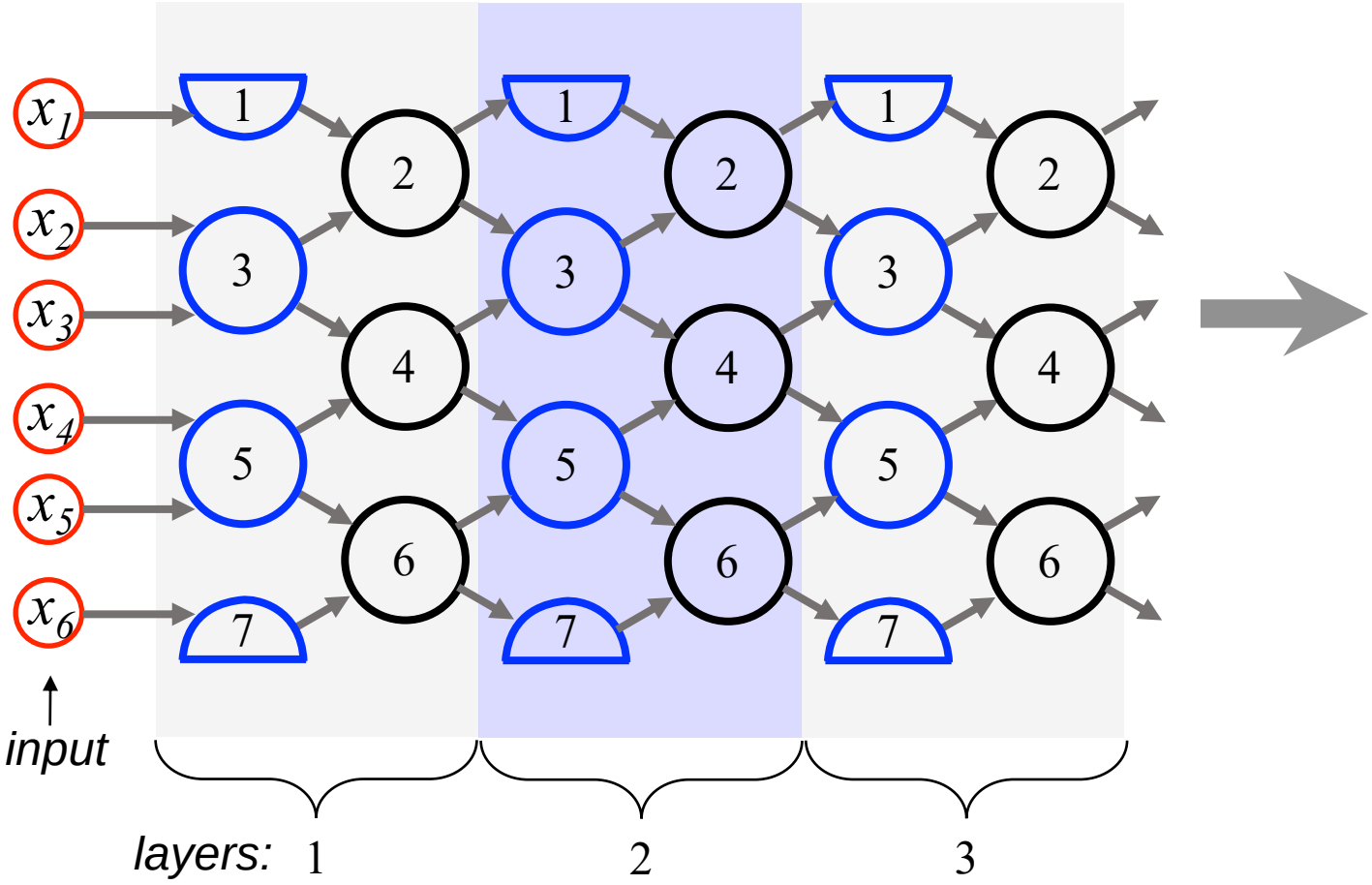
User gives n input bits



Circuit model

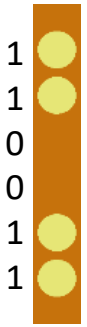
Programmer specifies layer: gates to go in each row

User gives n input bits

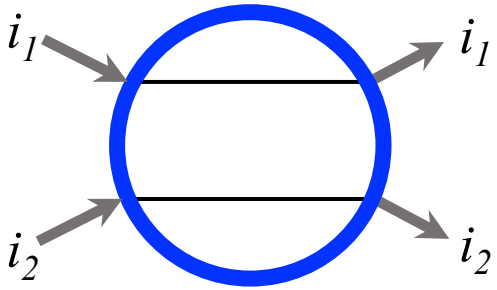


Example circuits with same gate in every row

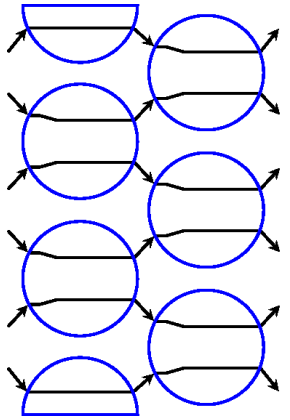
COPY



COPY gates

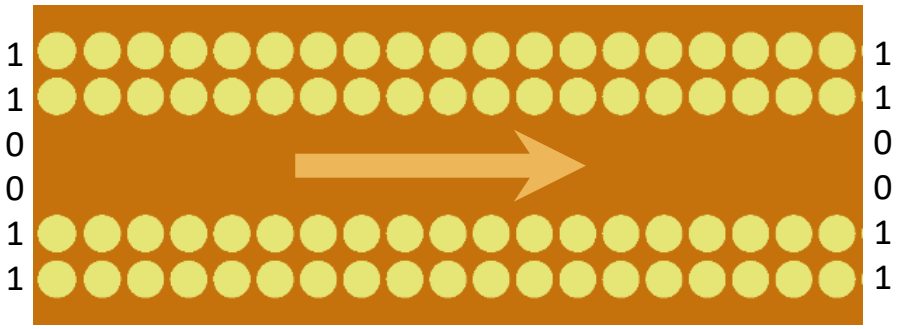


i_1	i_2	o_1	o_2
0	0	0	0
0	1	0	1
1	0	1	0
1	1	1	1

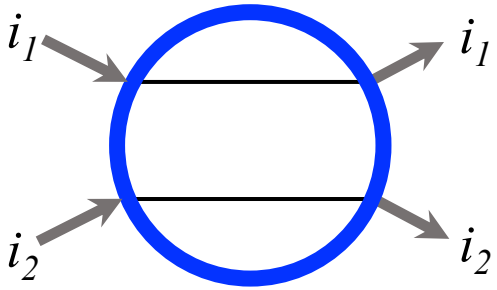


Example circuits with same gate in every row

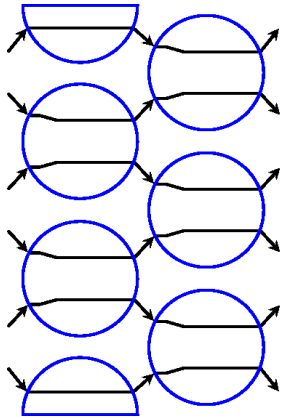
COPY



COPY gates

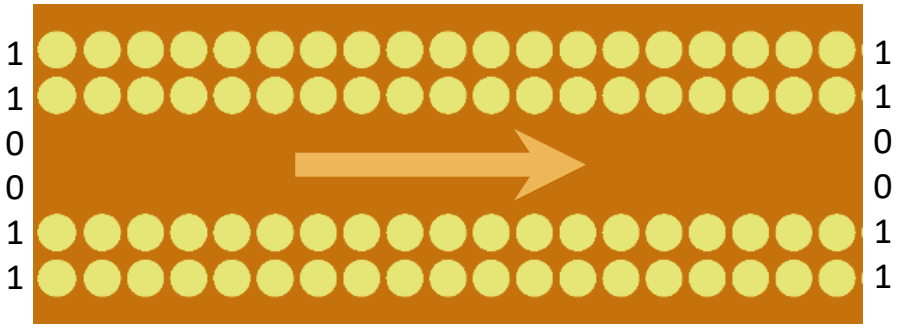


i_1	i_2	o_1	o_2
0	0	0	0
0	1	0	1
1	0	1	0
1	1	1	1

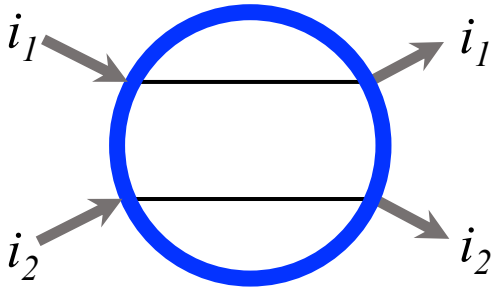


Example circuits with same gate in every row

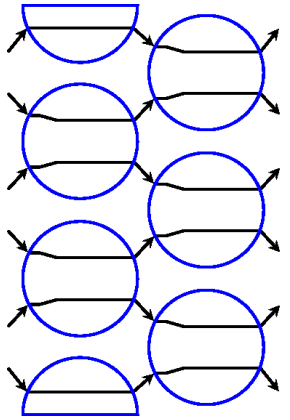
COPY



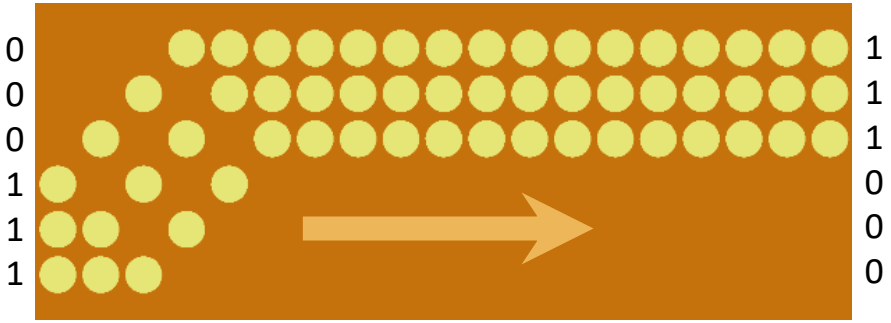
COPY gates



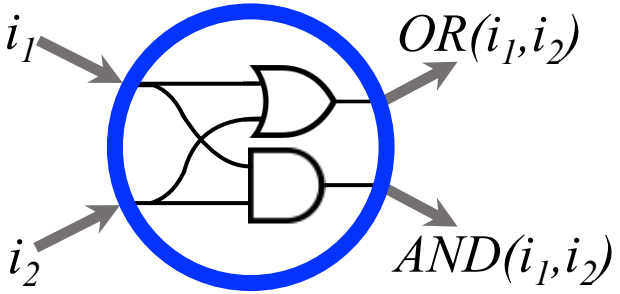
i_1	i_2	o_1	o_2
0	0	0	0
0	1	0	1
1	0	1	0
1	1	1	1



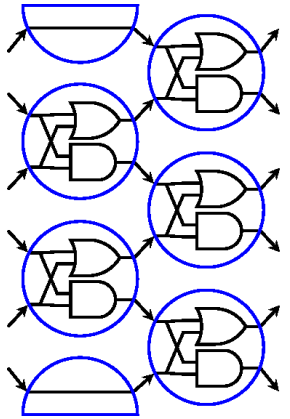
SORTING



SORTING gates

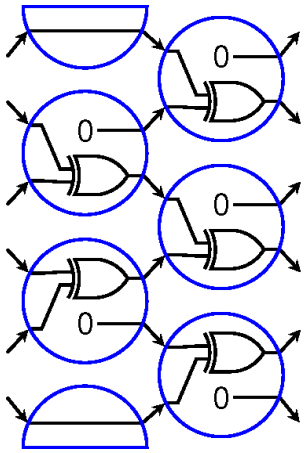


i_1	i_2	o_1	o_2
0	0	0	0
0	1	1	0
1	0	1	0
1	1	1	1



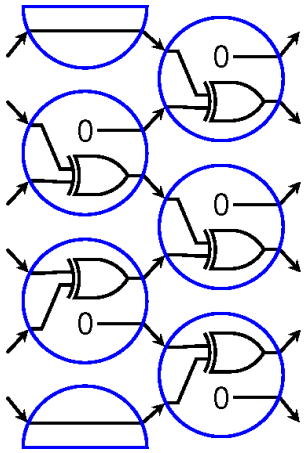
Example circuits with different gates in each row

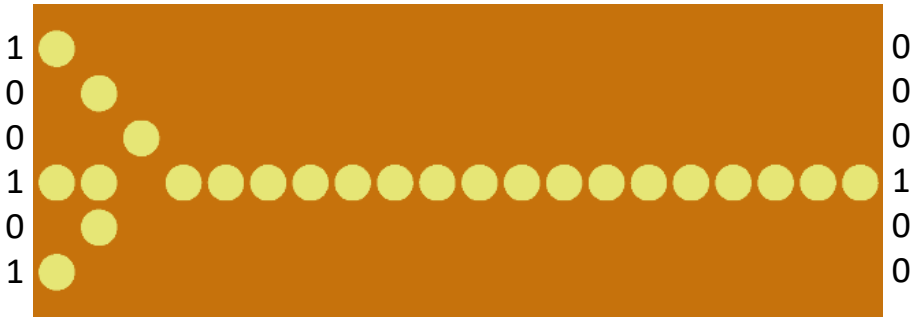
PARITY



Example circuits with different gates in each row

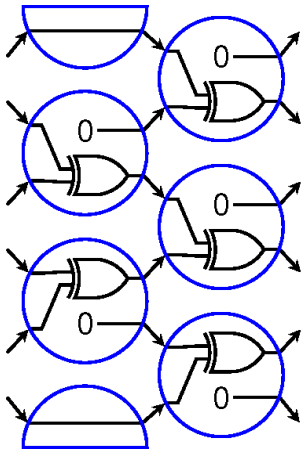
PARITY

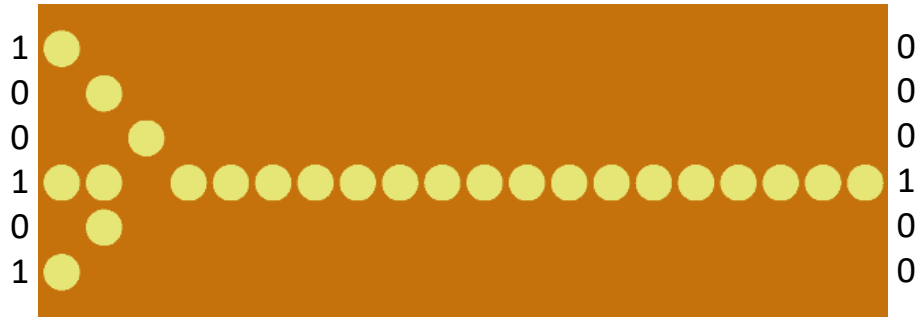




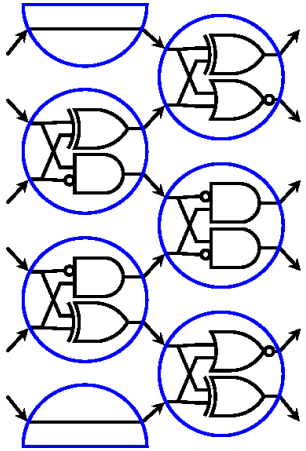
Example circuits with different gates in each row

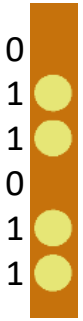
PARITY





MULTIPLEOF3

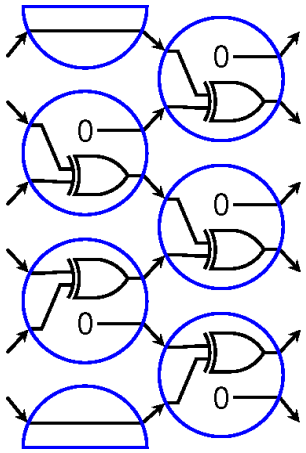


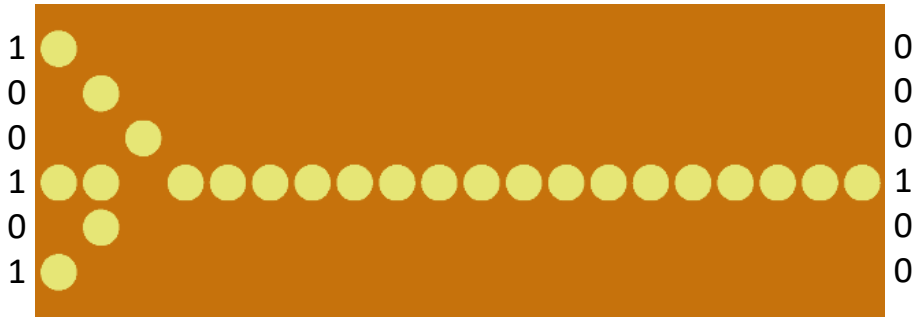


011011_2

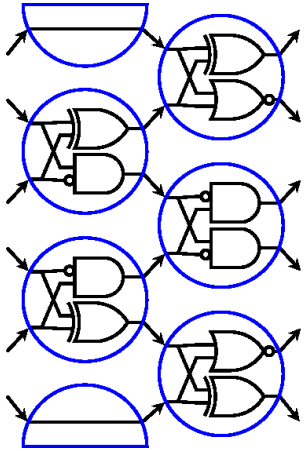
Example circuits with different gates in each row

Parity

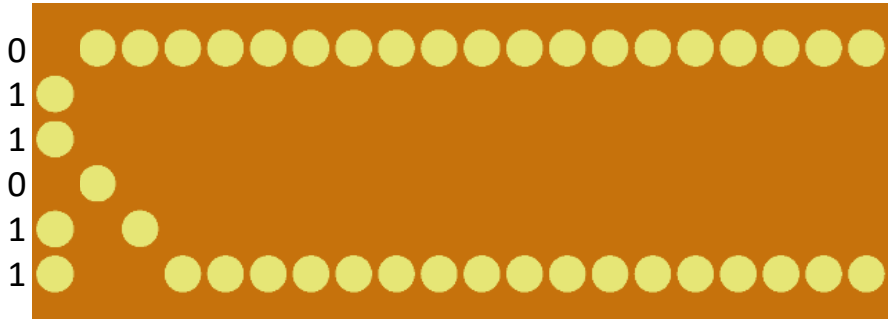




MultipleOf3

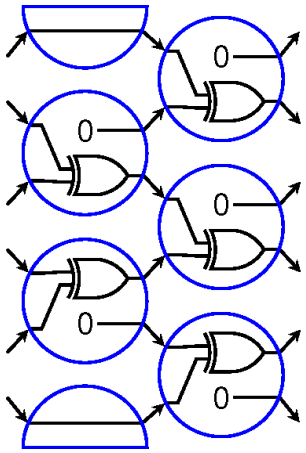


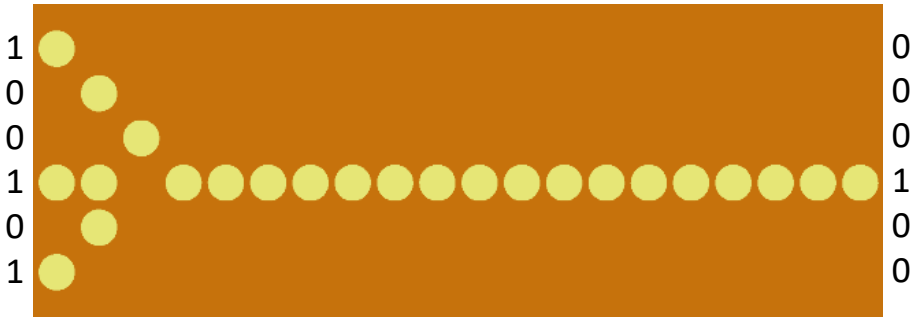
$011011_2 = 27_{10} = 3 \cdot 9$



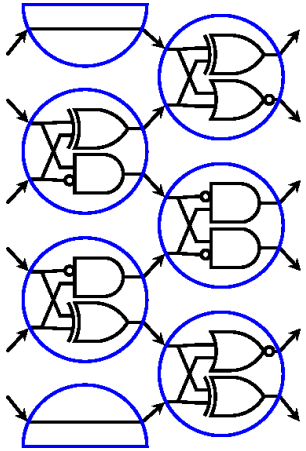
Example circuits with different gates in each row

PARITY

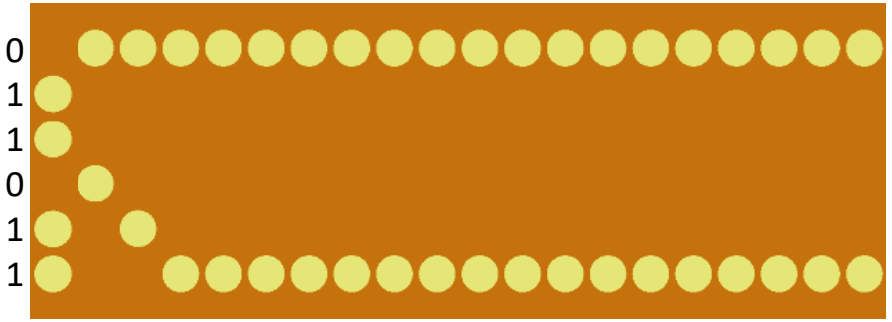




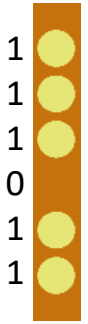
MULTIPLEOF3



$011011_2 = 27_{10} = 3 \cdot 9$

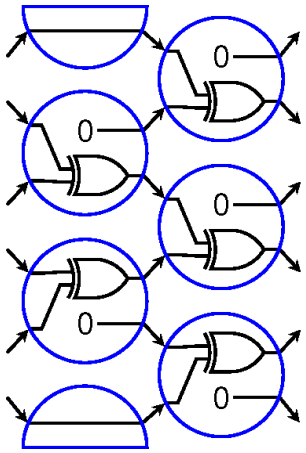


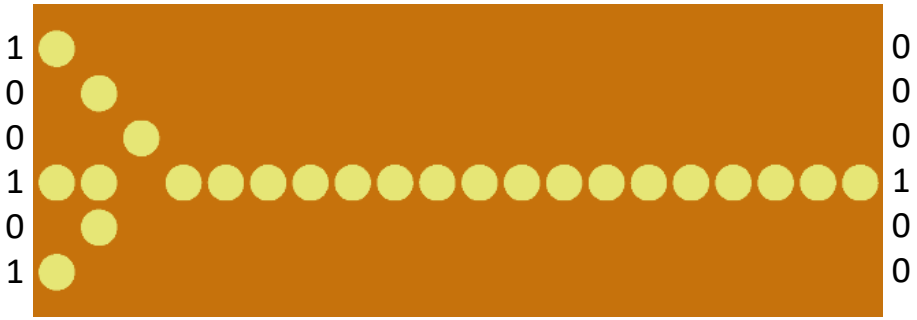
111011_2



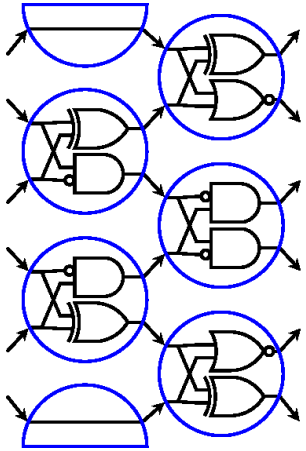
Example circuits with different gates in each row

Parity

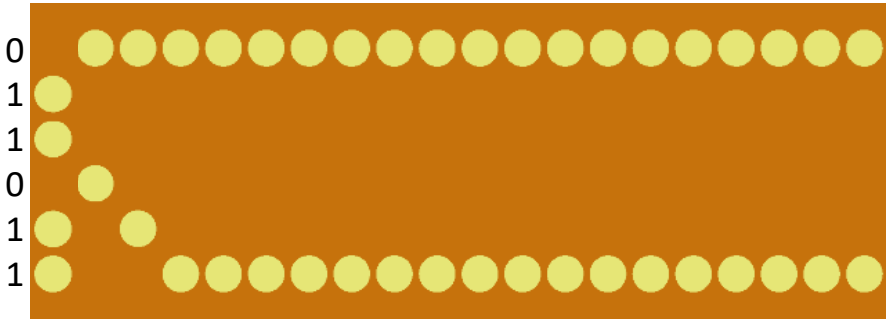




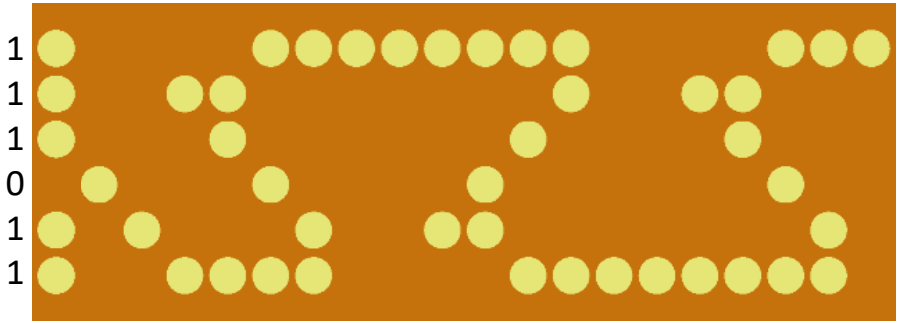
MultipleOf3



$011011_2 = 27_{10} = 3 \cdot 9$

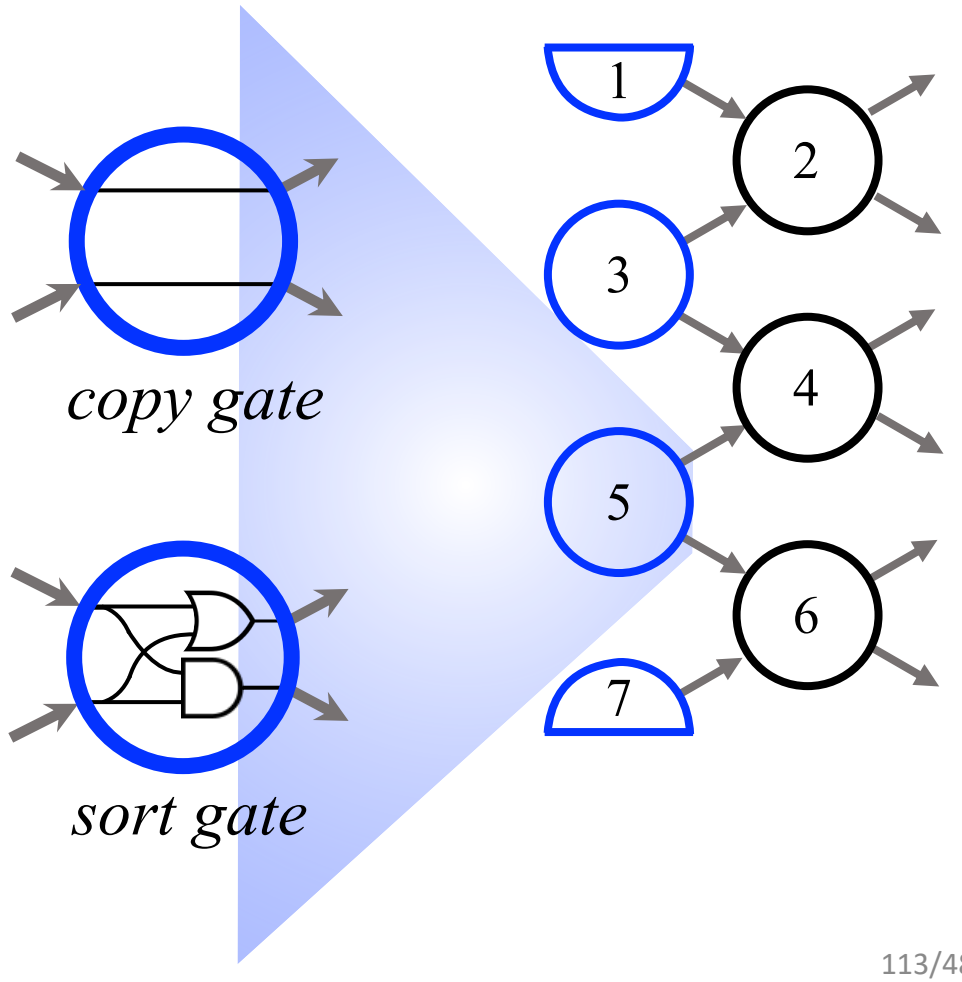


$111011_2 = 59_{10} = 3 \cdot 19 + 2$

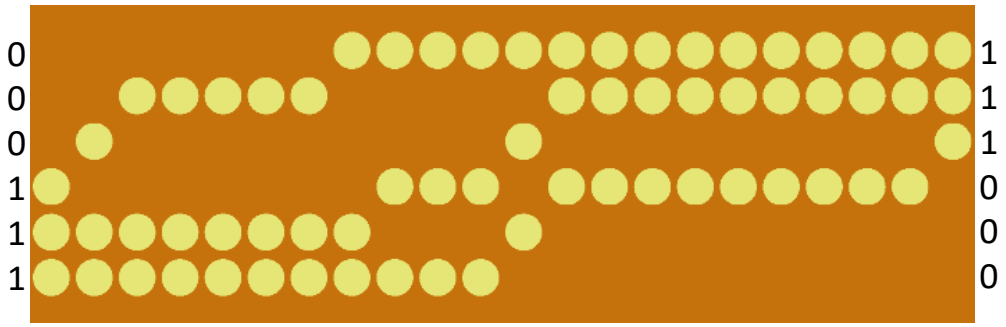


Randomization: “Lazy” sorting

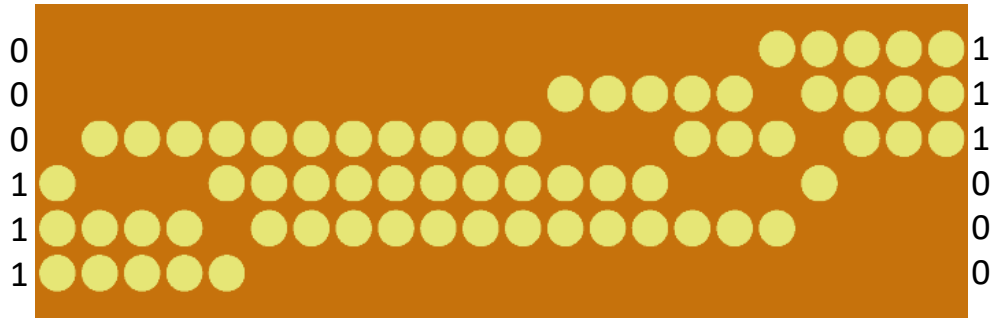
If 1 and 0 out of order, flip a coin to decide whether to swap them.

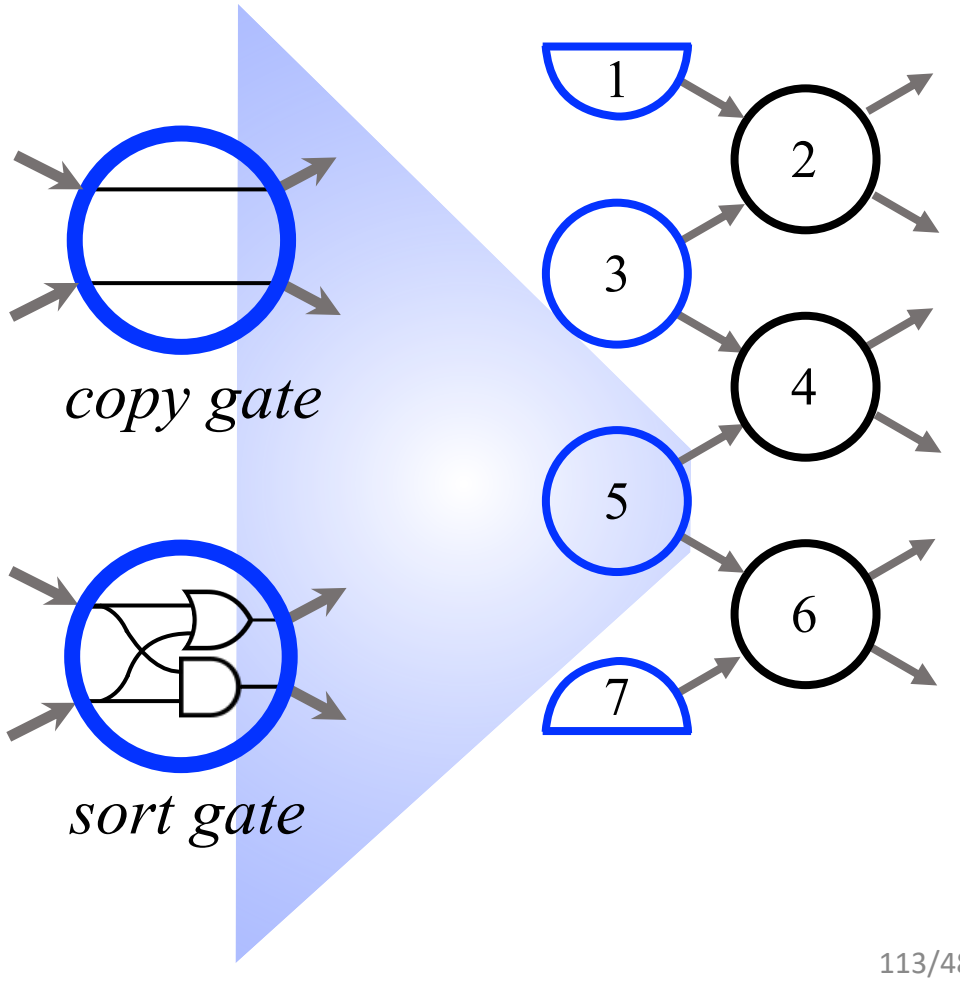


Randomization: “Lazy” sorting



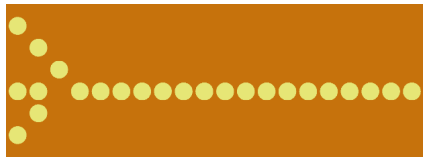
If 1 and 0 out of order, flip a coin to decide whether to swap them.





Deterministic circuits

PARITY



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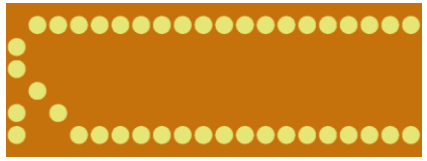
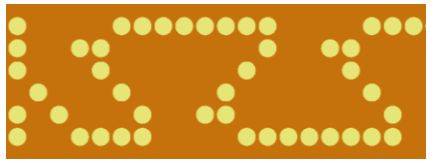
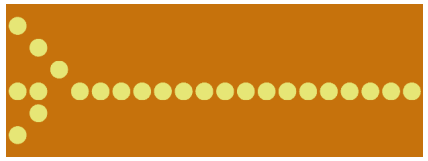




answer yes/no question

Deterministic circuits

PARITY



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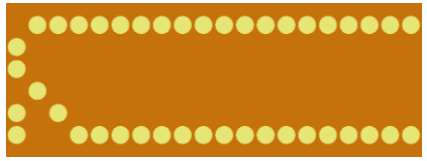
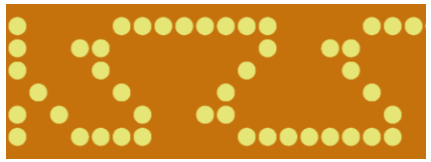




PALINDROME

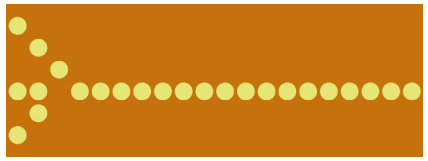
yes

no

answer yes/no question

Deterministic circuits

PARITY



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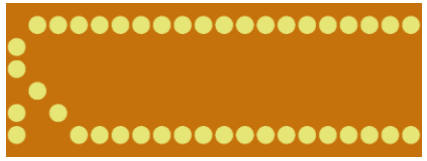
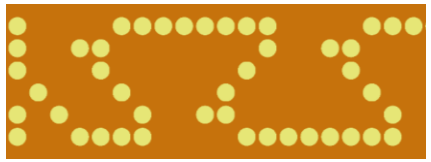


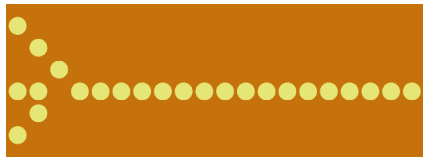


PALINDROME

answer yes/no question

Deterministic circuits

PARITY



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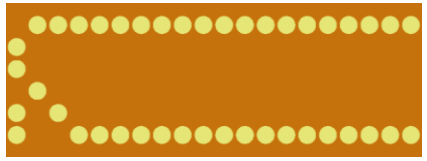
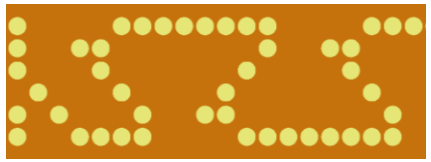


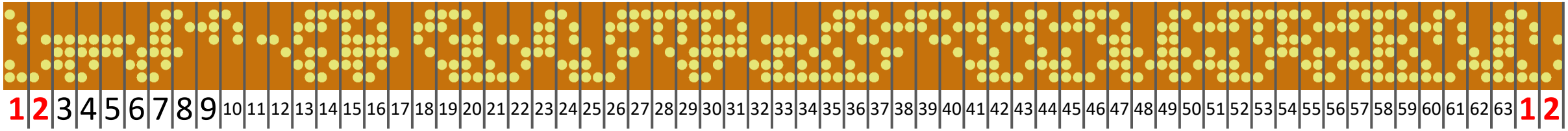


PALINDROME

answer yes/no question

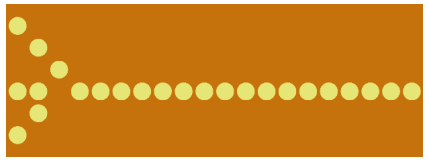
CYCLE63

"count" as high as possible

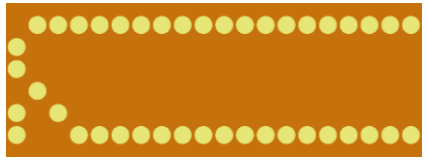


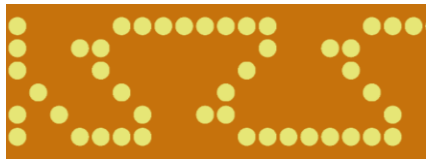
Deterministic circuits

PARITY



MULTIPLEOF3



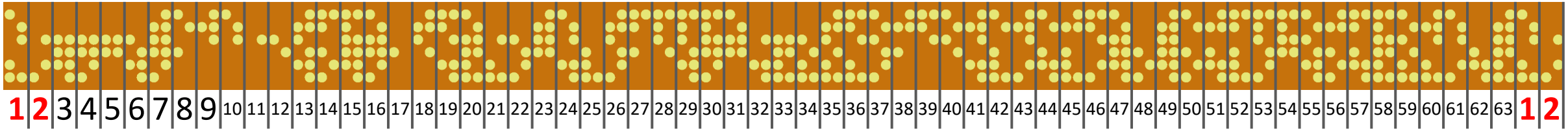


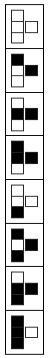
PALINDROME

answer yes/no question

CYCLE63

"count" as high as possible



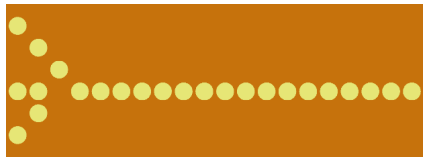


RULE110

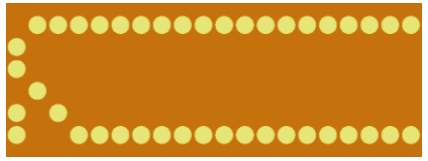
simulate cellular automata

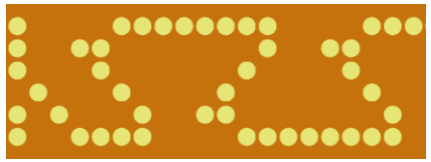
Deterministic circuits

PARITY

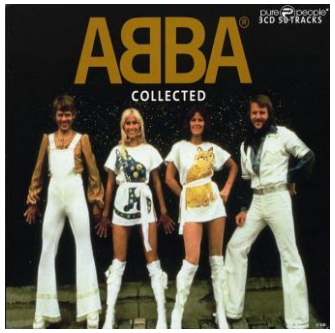


MULTIPLEOF3





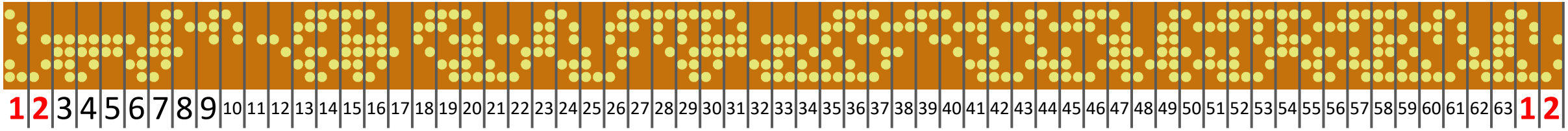
PALINDROME



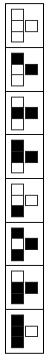
answer yes/no question

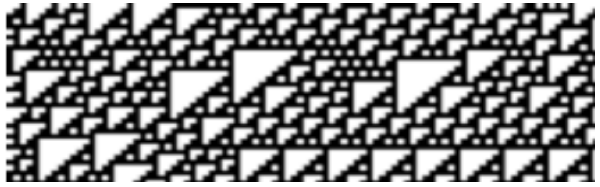
CYCLE63

"count" as high as possible



RULE110



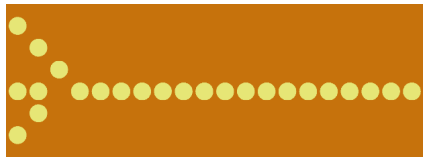


time

simulate cellular automata

Deterministic circuits

PARITY



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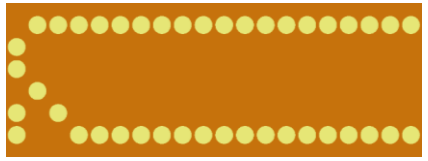
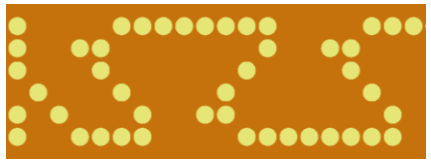


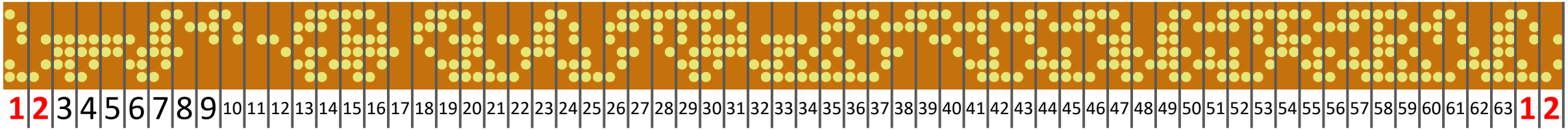


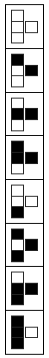
PALINDROME

answer yes/no question

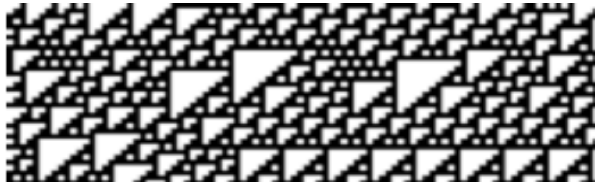
CYCLE63

"count" as high as possible

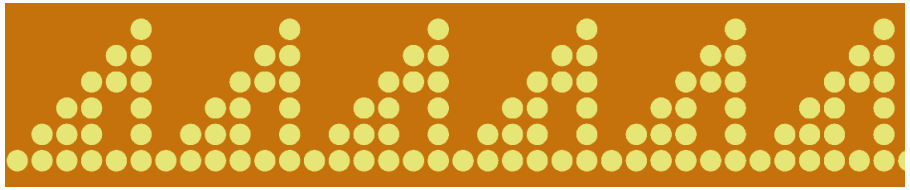




RULE110



time



simulate cellular automata

Theorem: Rule 110 can efficiently execute any algorithm.

[Cook, Complex Systems 2004]

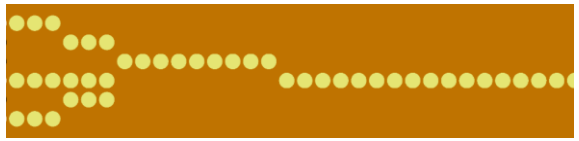
[Neary, Woods, ICALP 2006]

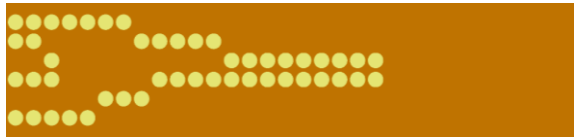
Randomized circuits

LAZYPARITY

Randomized circuits

LAZYPARITY





Randomized circuits

LAZYPARITY

RANDOMWALKINGBIT

Randomized circuits

LAZYPARITY

RANDOMWALKINGBIT

DIAMONDSAREFOREVER

Randomized circuits

LAZYPARITY

RANDOMWALKINGBIT

DIAMONDSAREFOREVER

FAIRCOIN

use biased coin to
simulate unbiased coin

Randomized circuits

LAZYPARITY

RANDOMWALKINGBIT

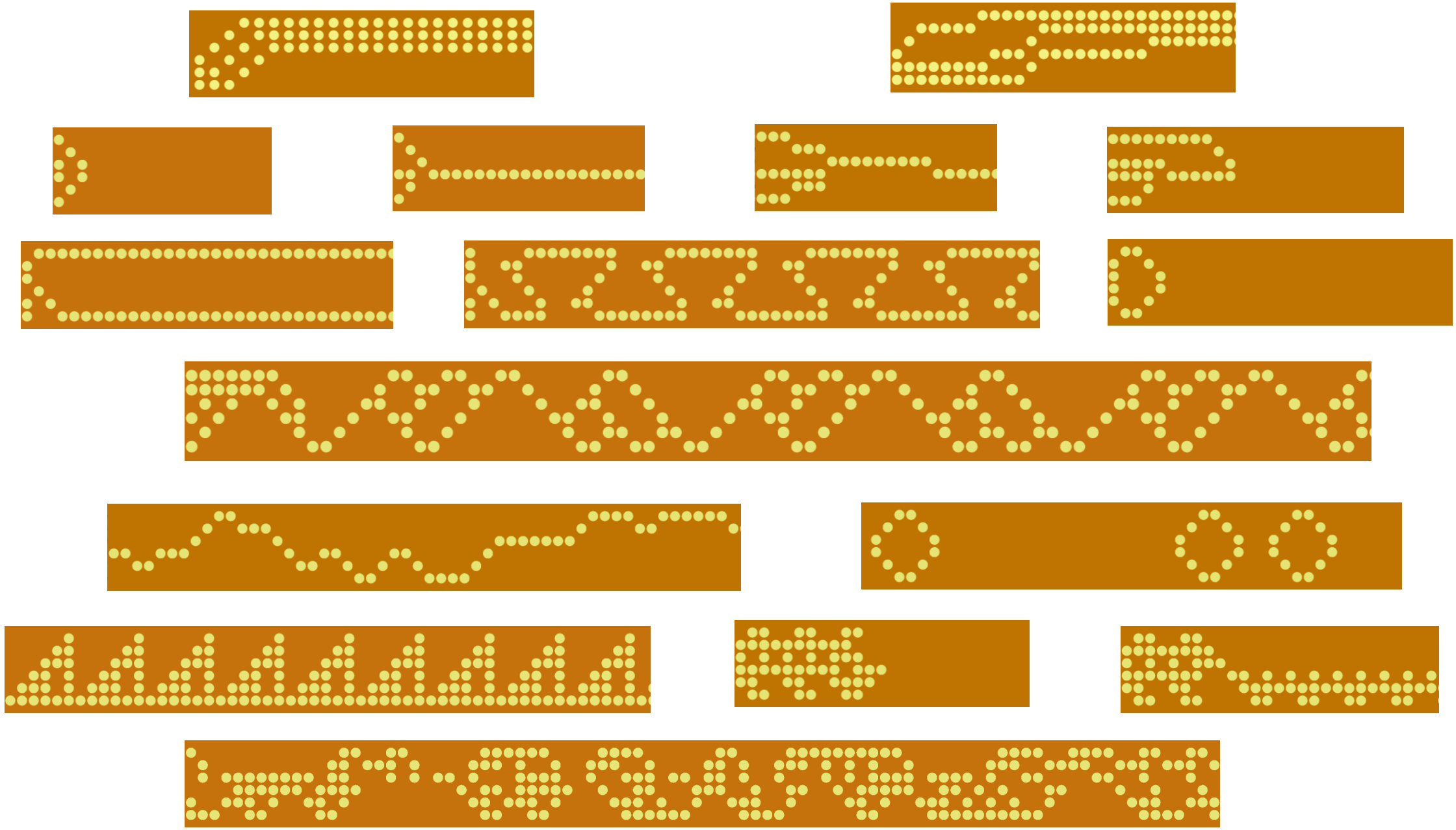
DIAMONDSAREFOREVER

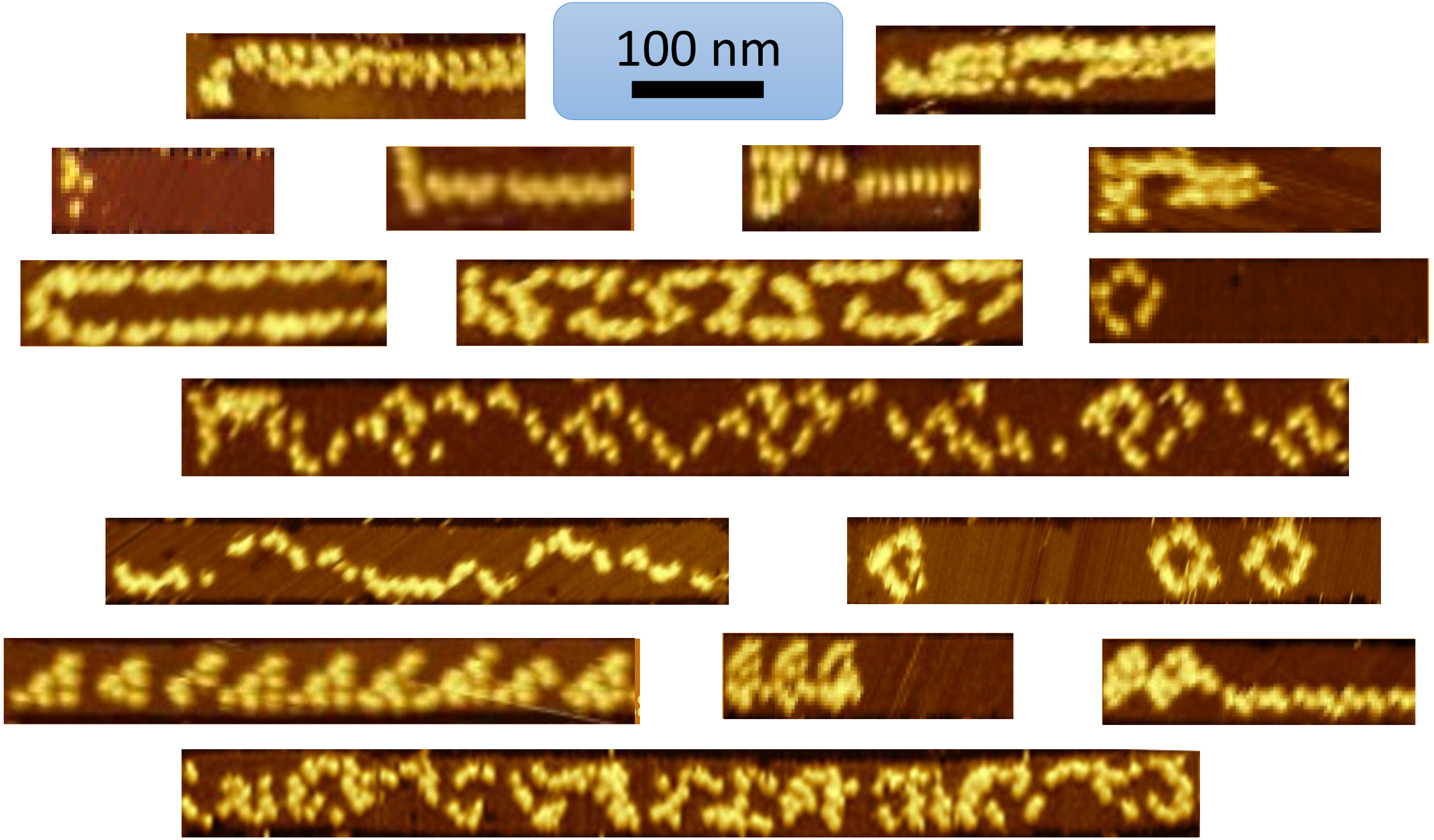
FAIRCOIN

use biased coin to
simulate unbiased coin

$$\Pr\left[\ \text{  }\ \right] = \Pr\left[\ \text{  }\ \right] = \tfrac{1}{2}$$

for **any** (positive) probabilities for the randomized gate




100 nm

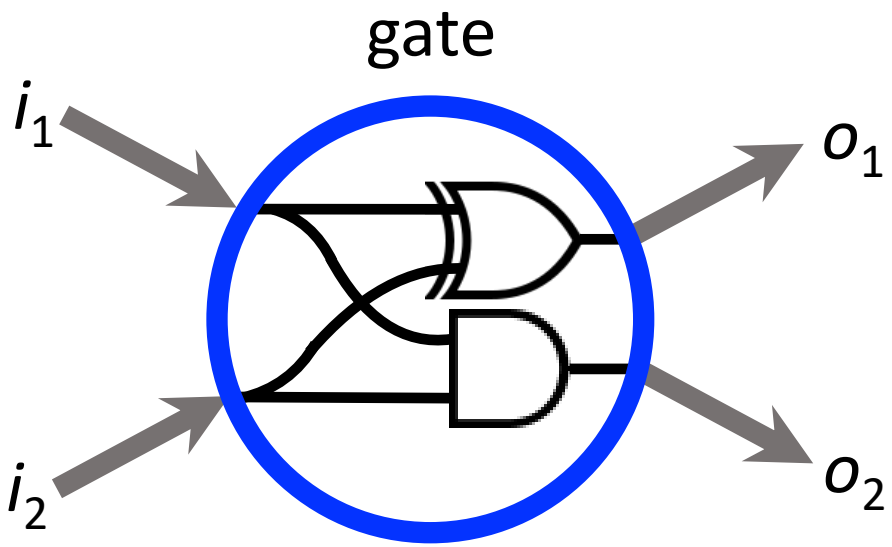
Hierarchy of abstractions

Bits: Boolean circuits compute

➡ **Tiles: Tile growth implements circuits**

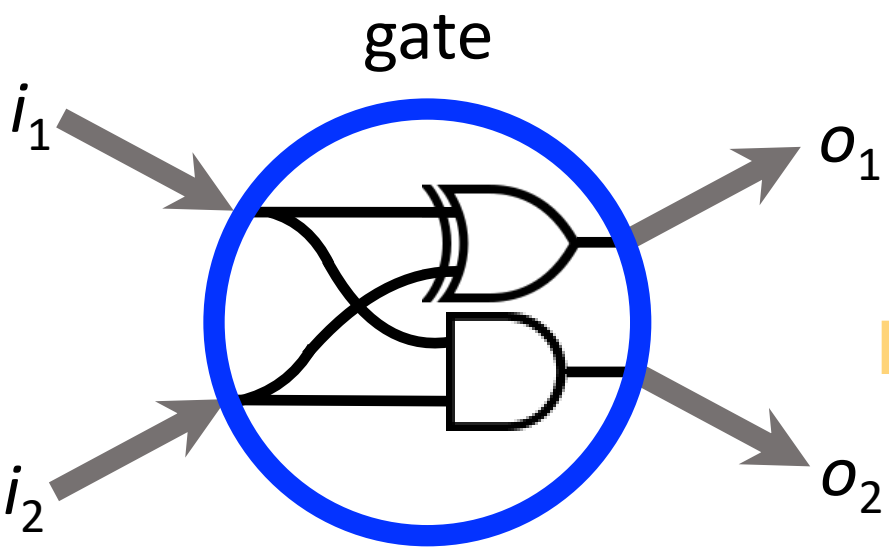
DNA: DNA strands implement tiles

Gates → Tiles

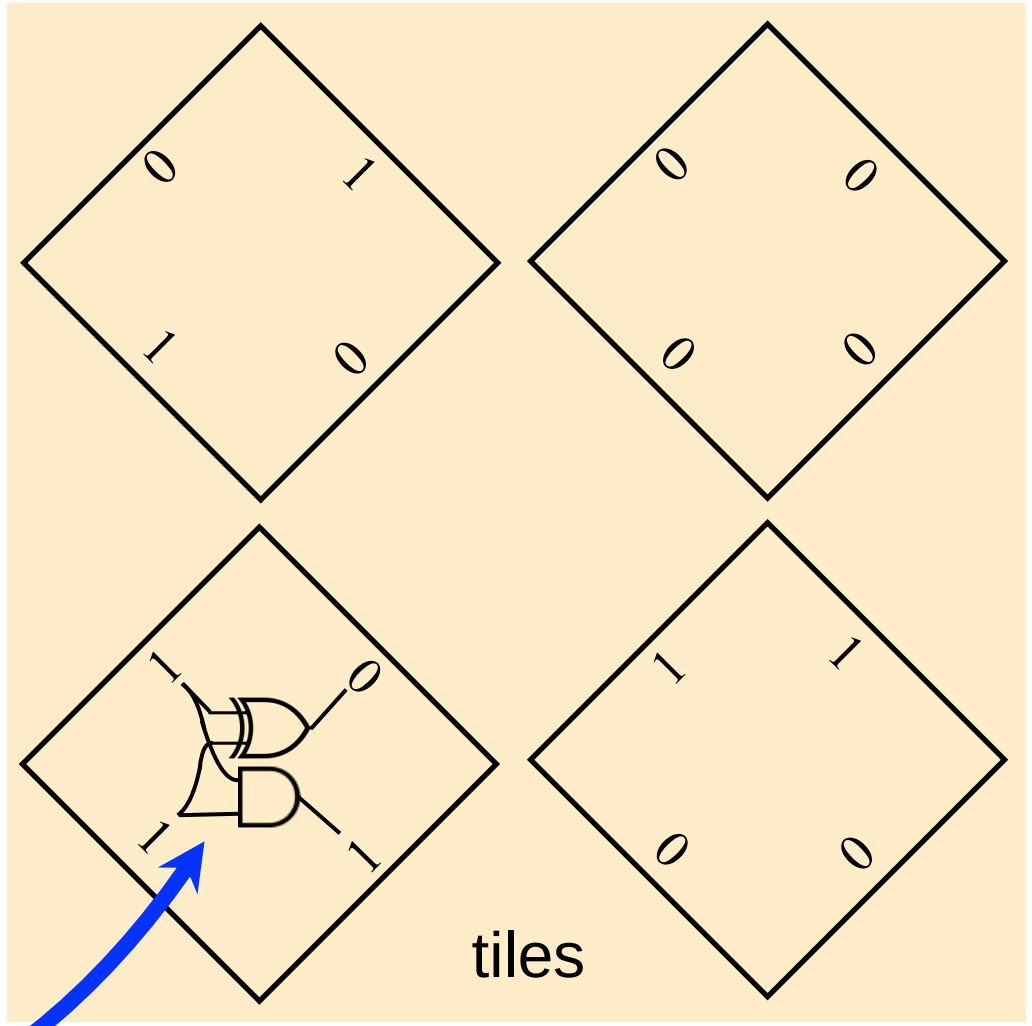


i_1	i_2	o_1	o_2
0	0	0	0
0	1	1	0
1	0	1	0
1	1	0	1

Gates → Tiles

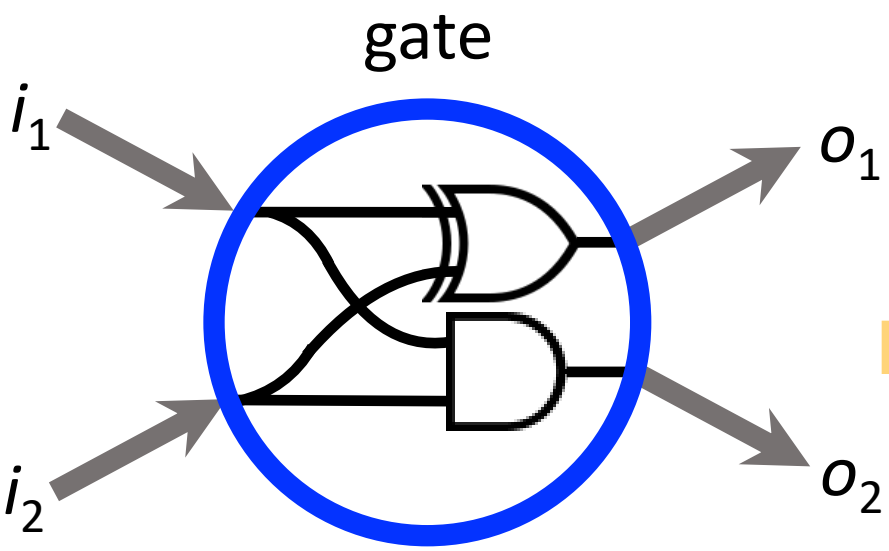


i_1	i_2	o_1	o_2
0	0	0	0
0	1	1	0
1	0	1	0
1	1	0	1

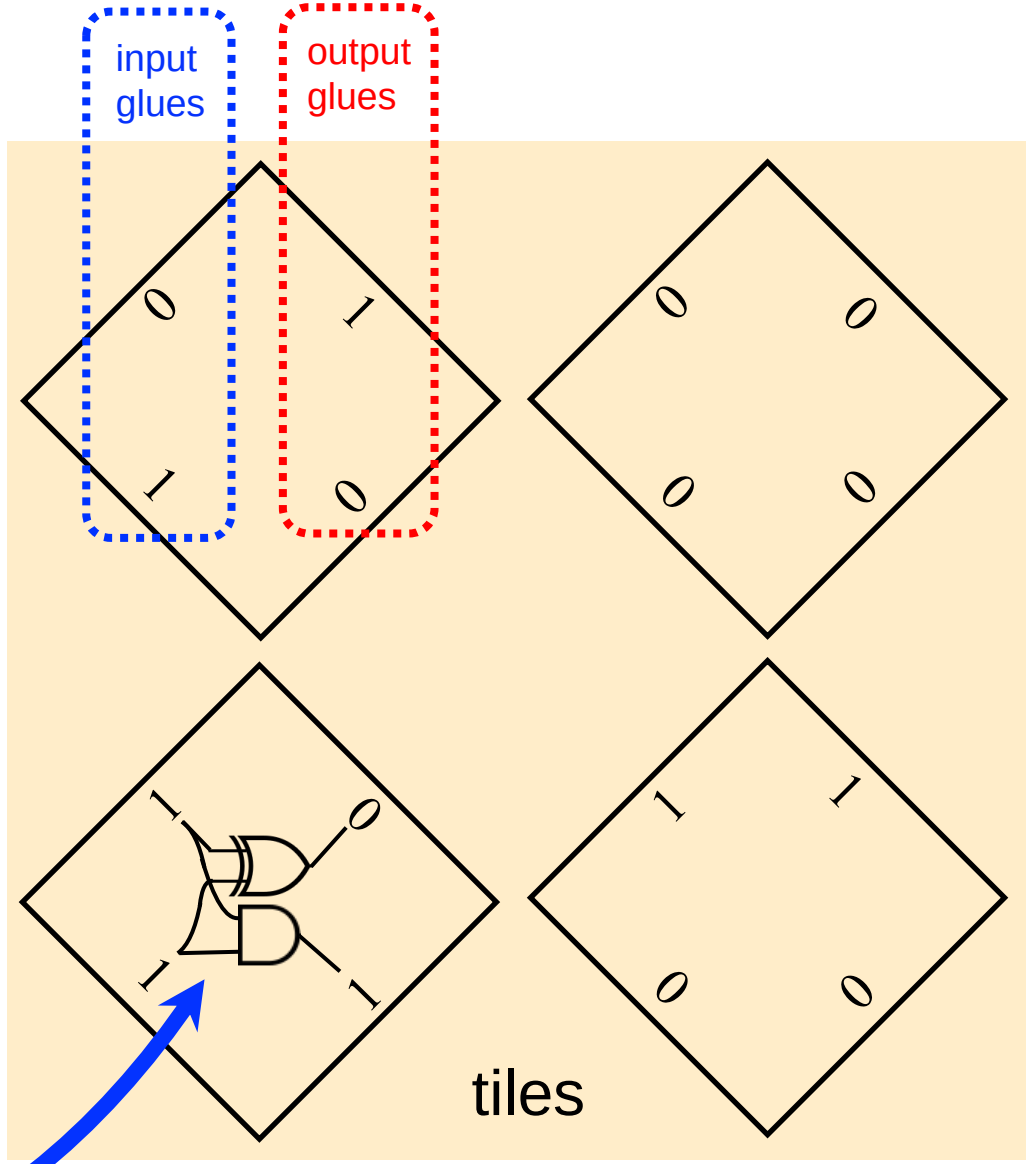


truth table row is encoded by a **tile** with 4 **glues** encoding bits

Gates → Tiles

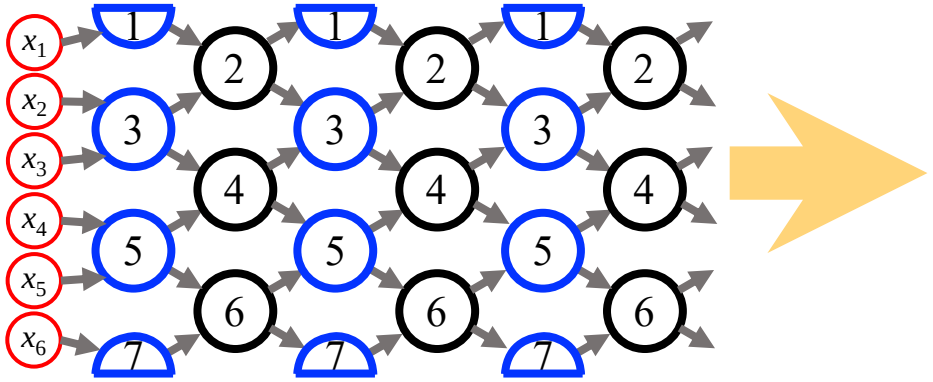


i_1	i_2	o_1	o_2
0	0	0	0
0	1	1	0
1	0	1	0
1	1	0	1



truth table row is encoded by a **tile** with 4 **glues** encoding bits

How tiles compute while growing (algorithmic self-assembly)



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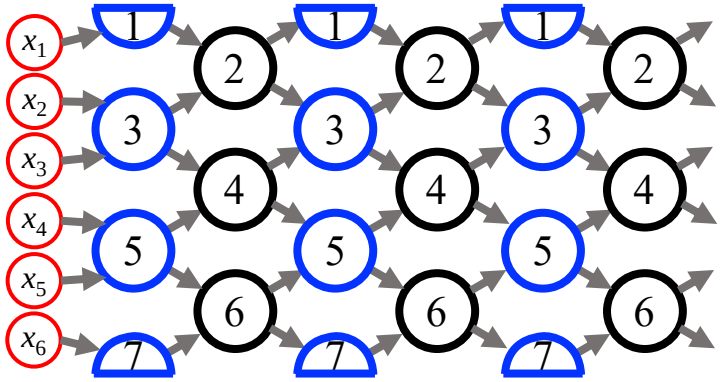

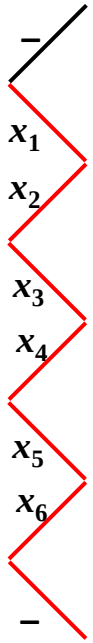

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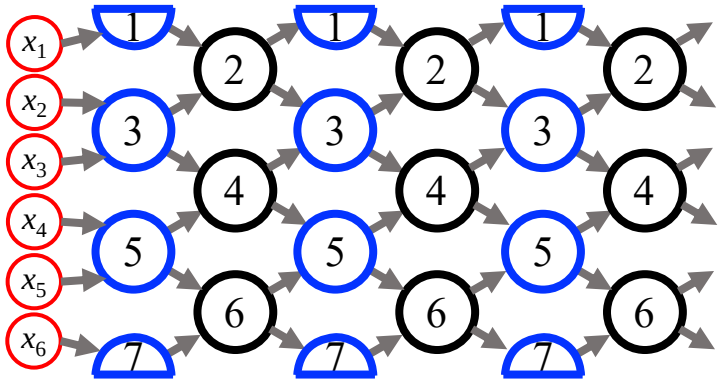

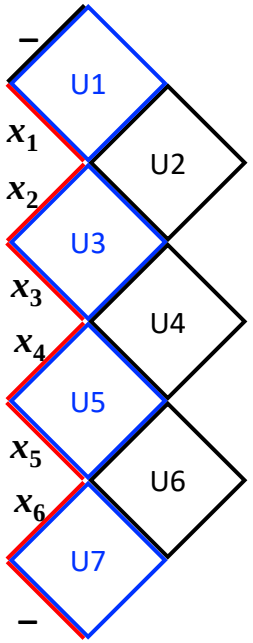

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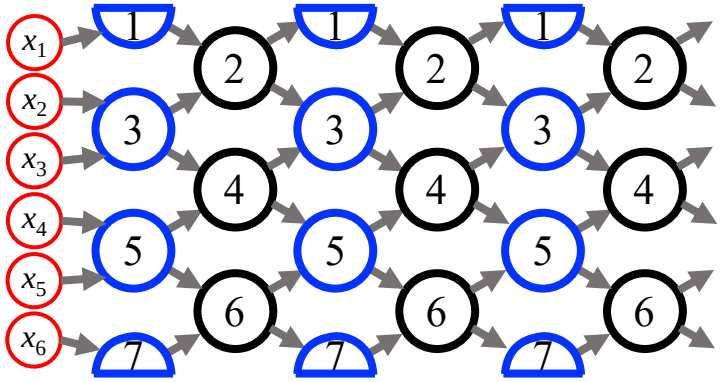
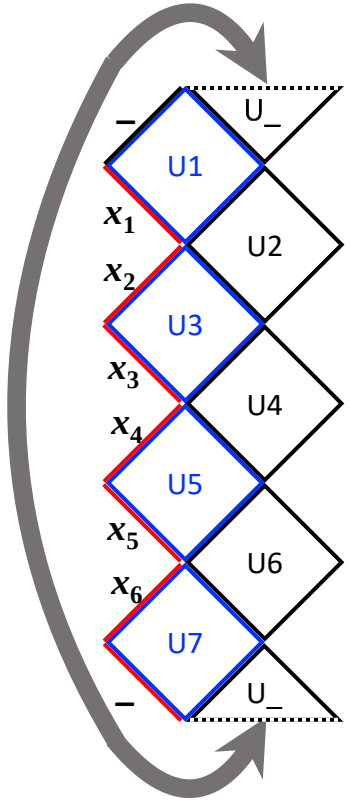
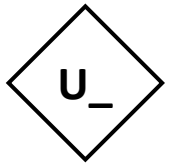




"data-free" tile wraps top to bottom to form a tube



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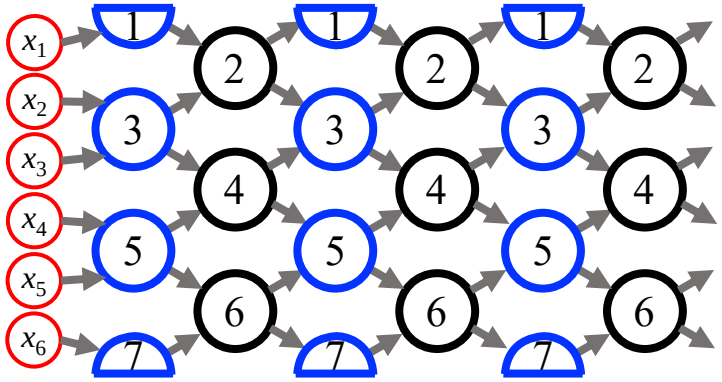

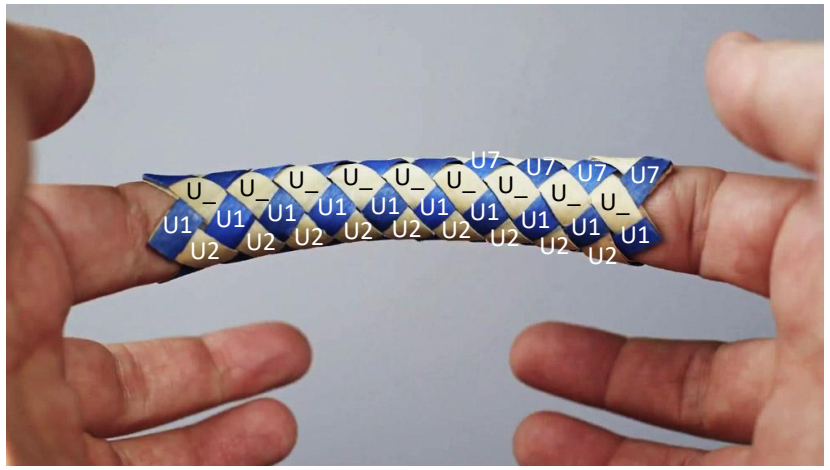

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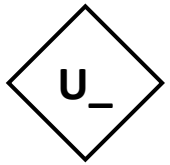

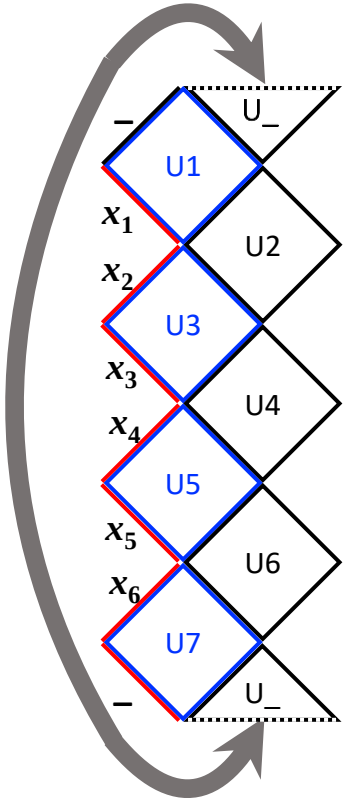

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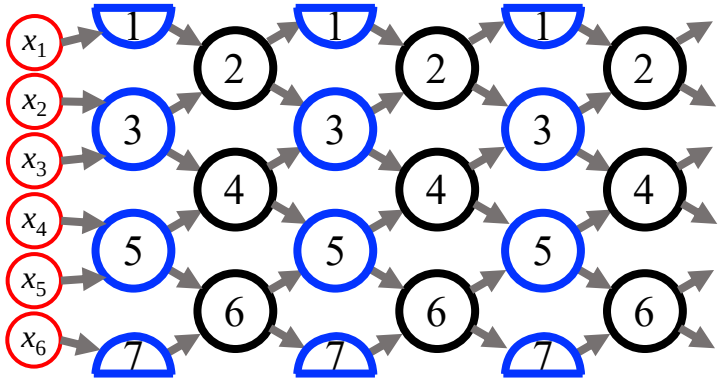

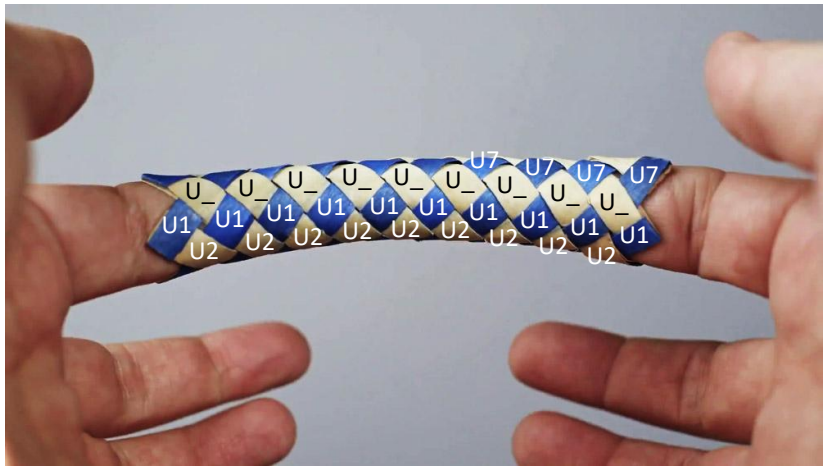

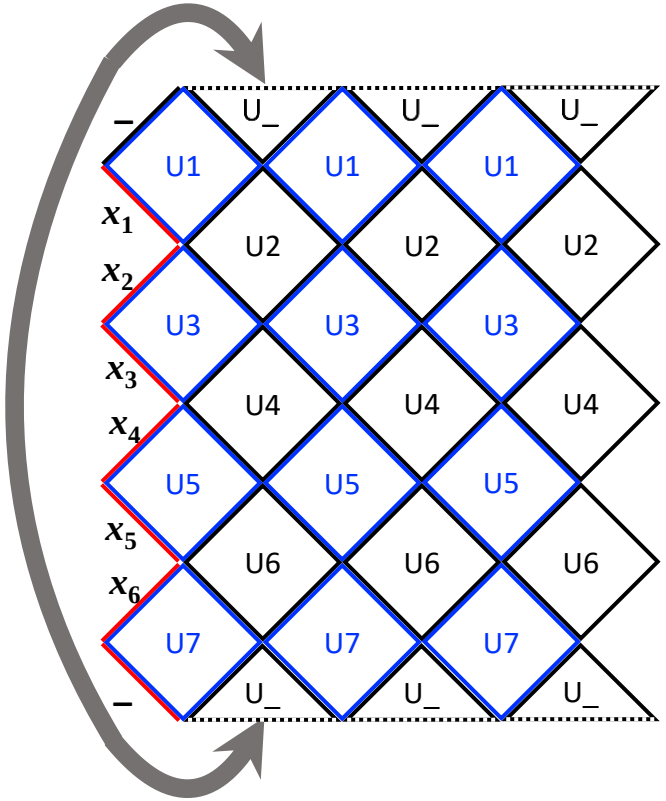

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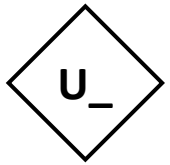

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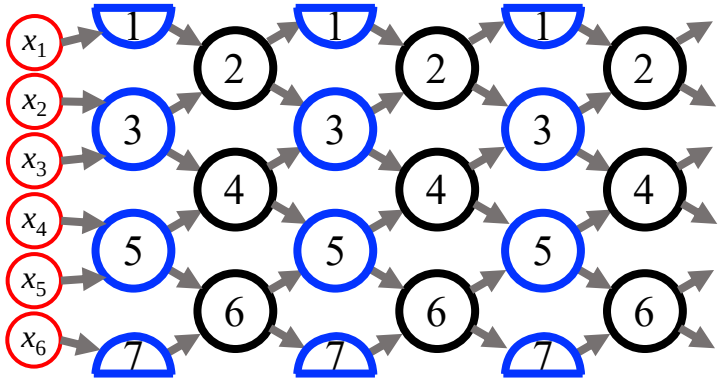

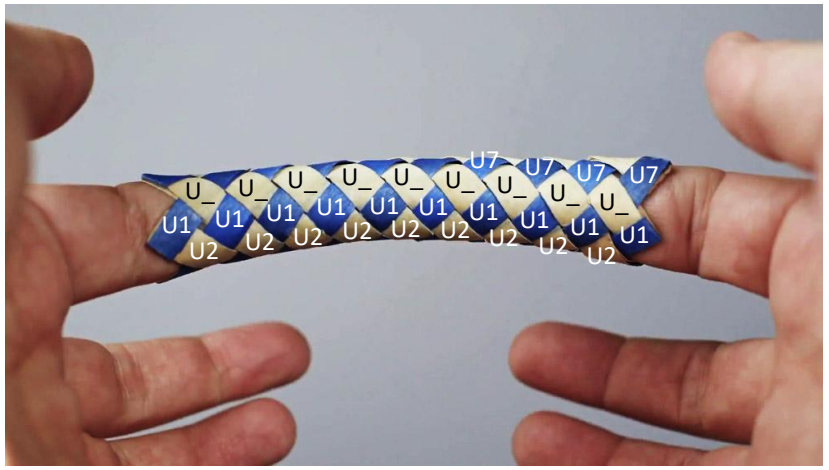

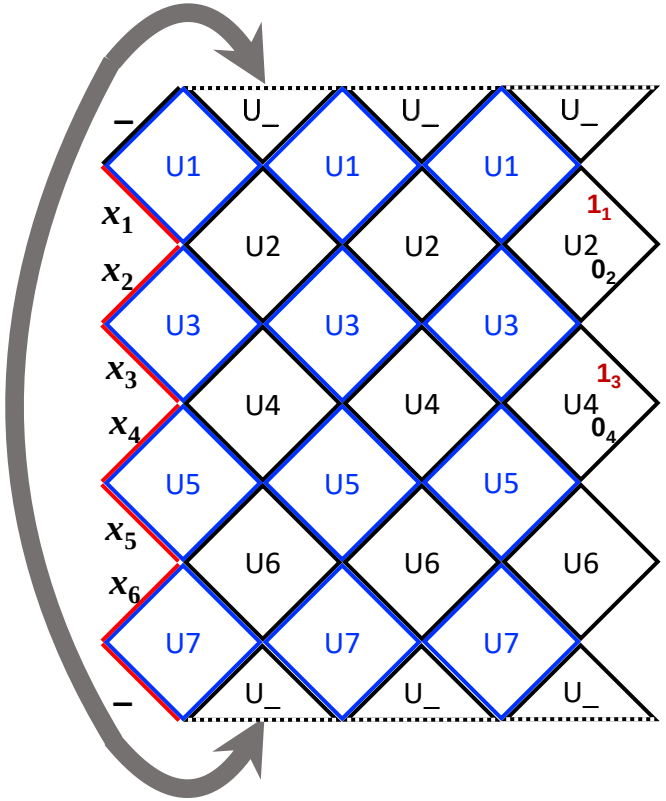

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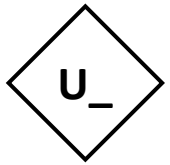

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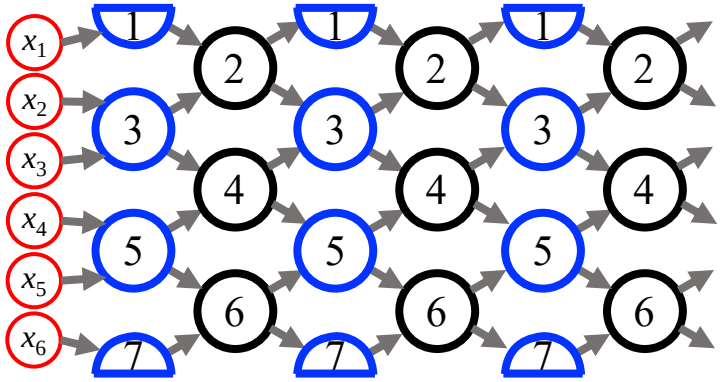

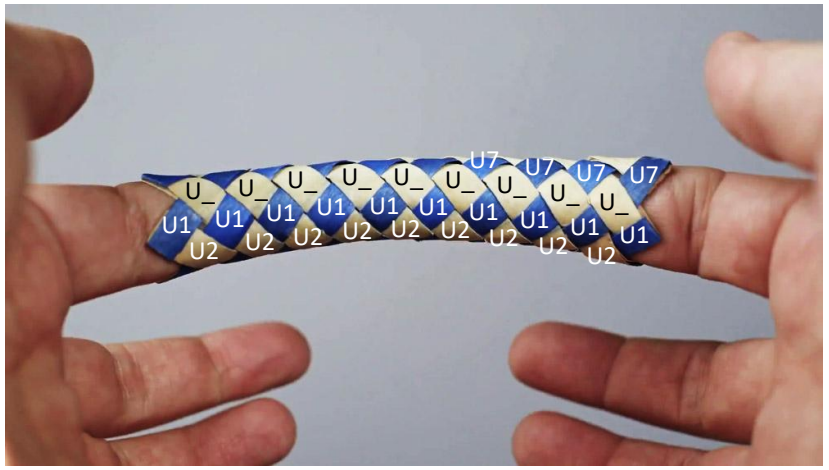

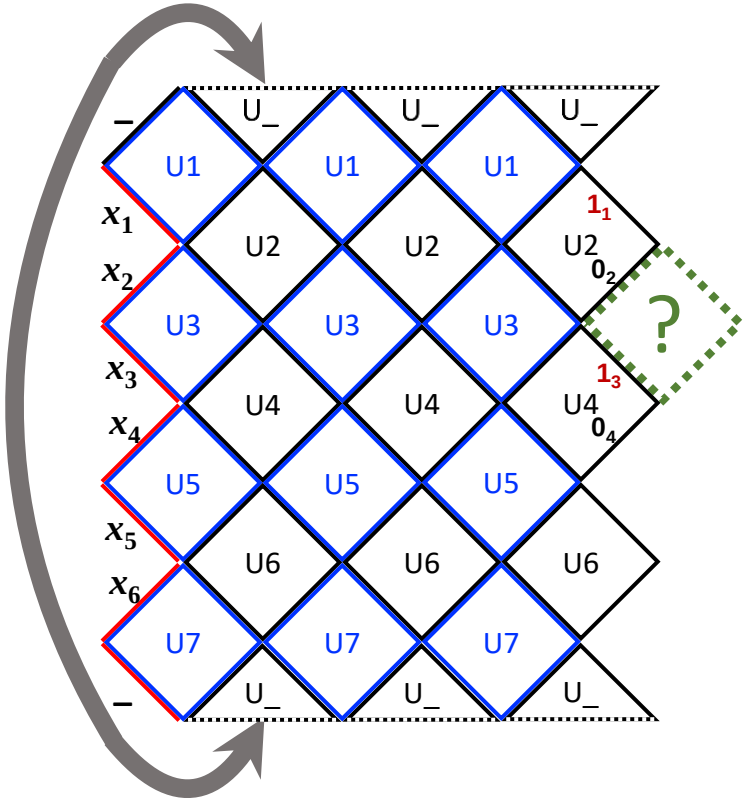

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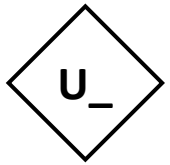

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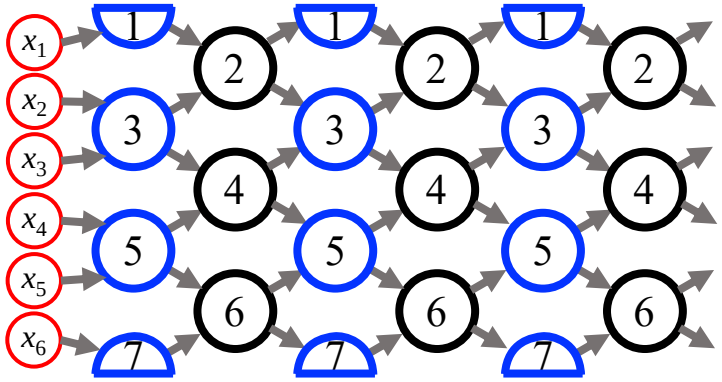

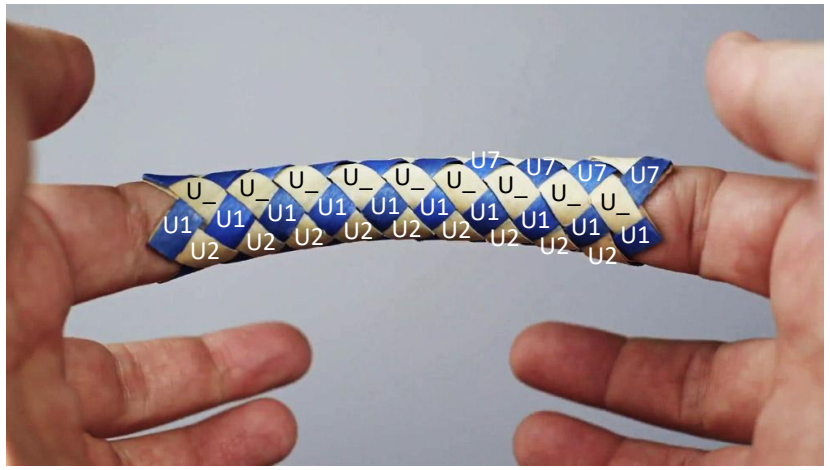

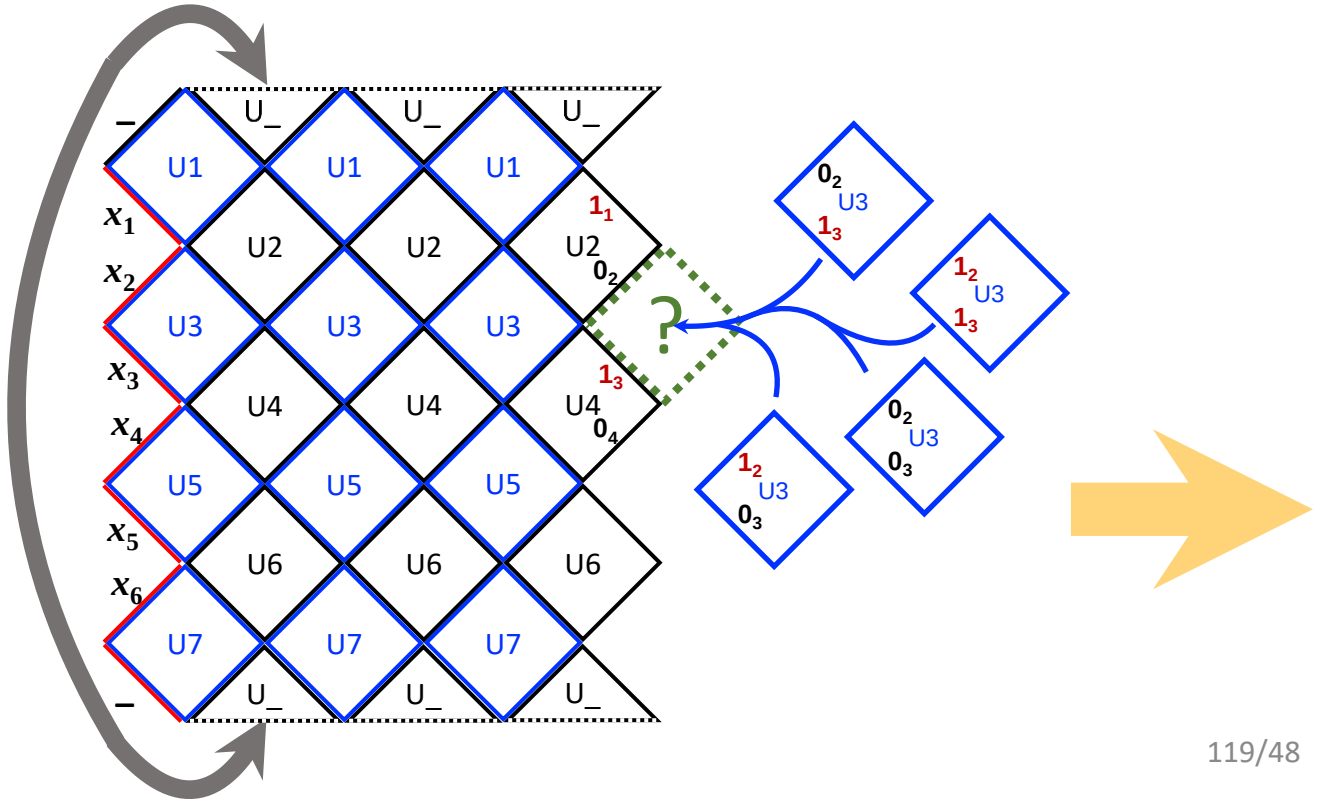

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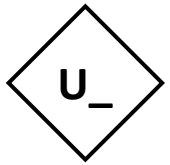

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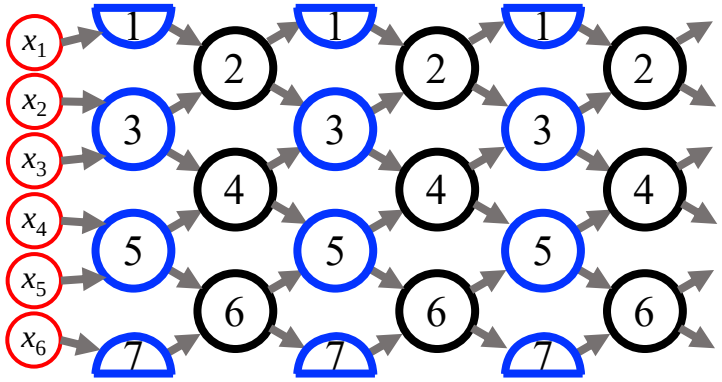

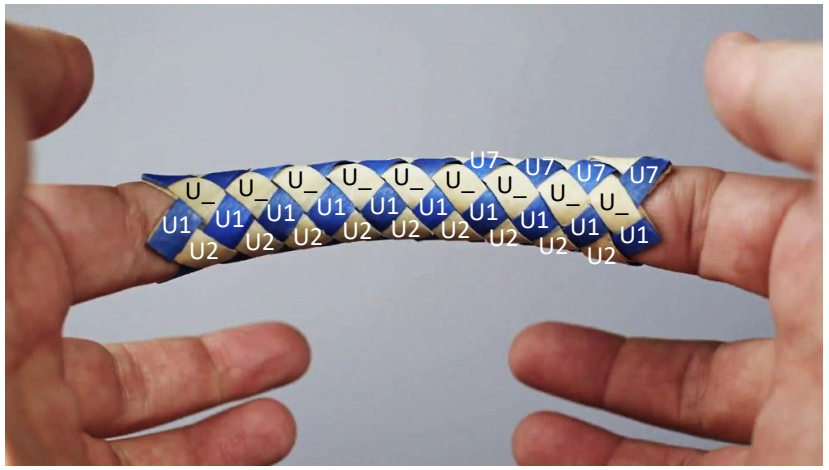

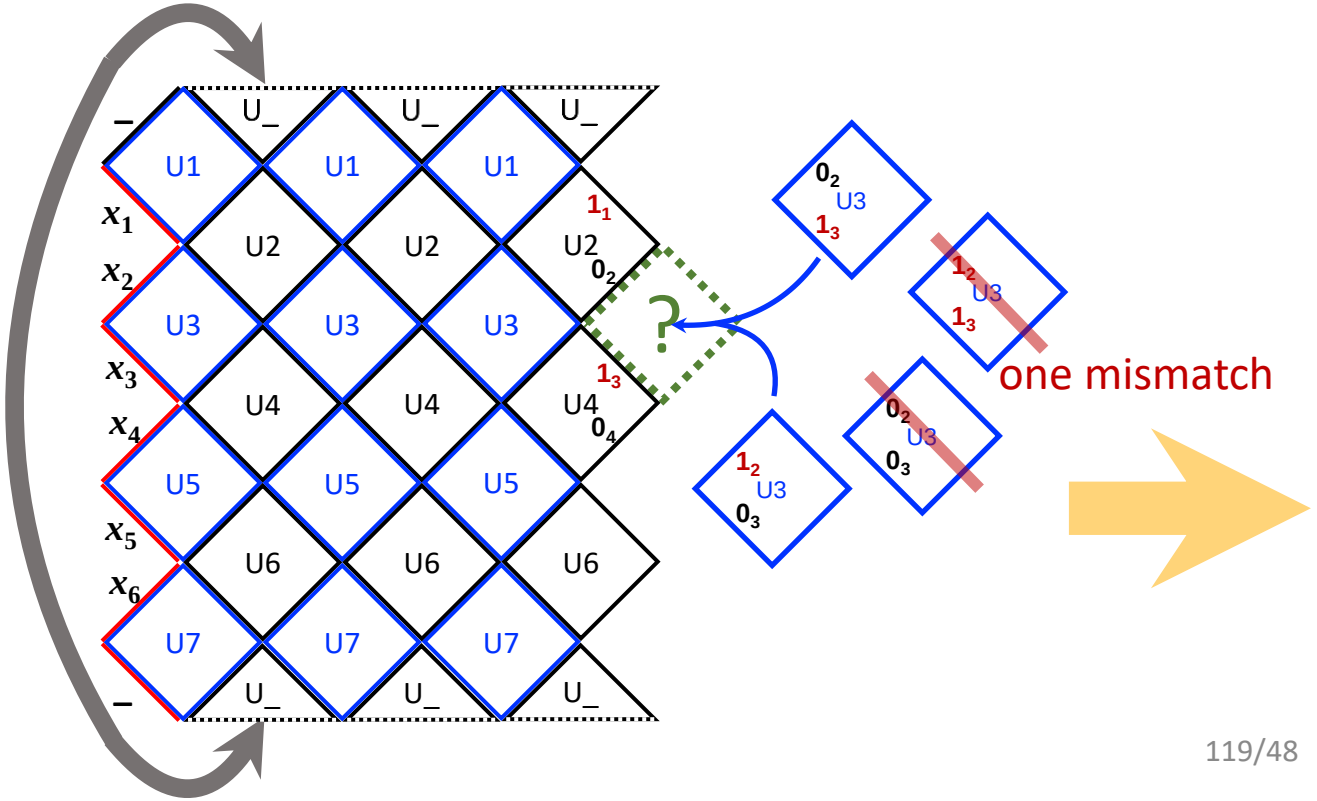

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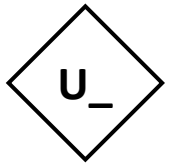

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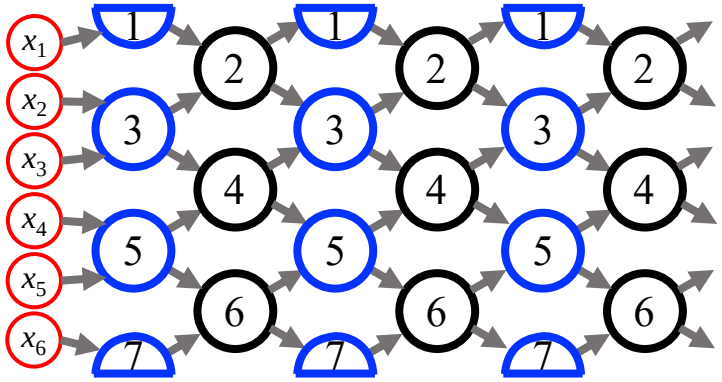

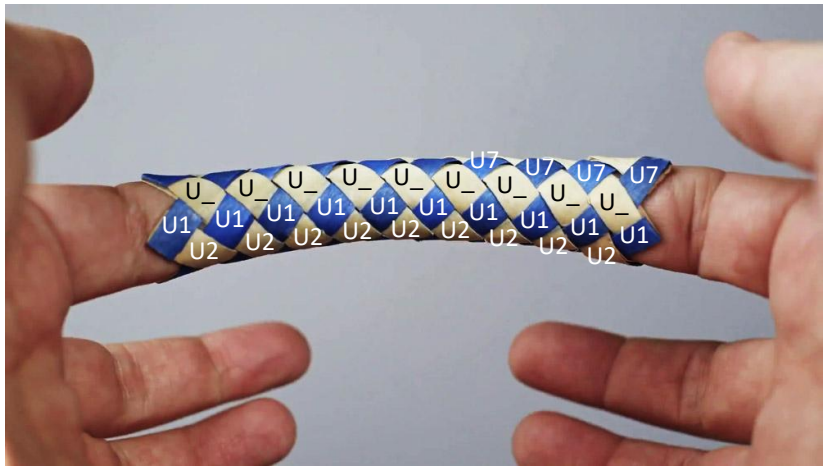

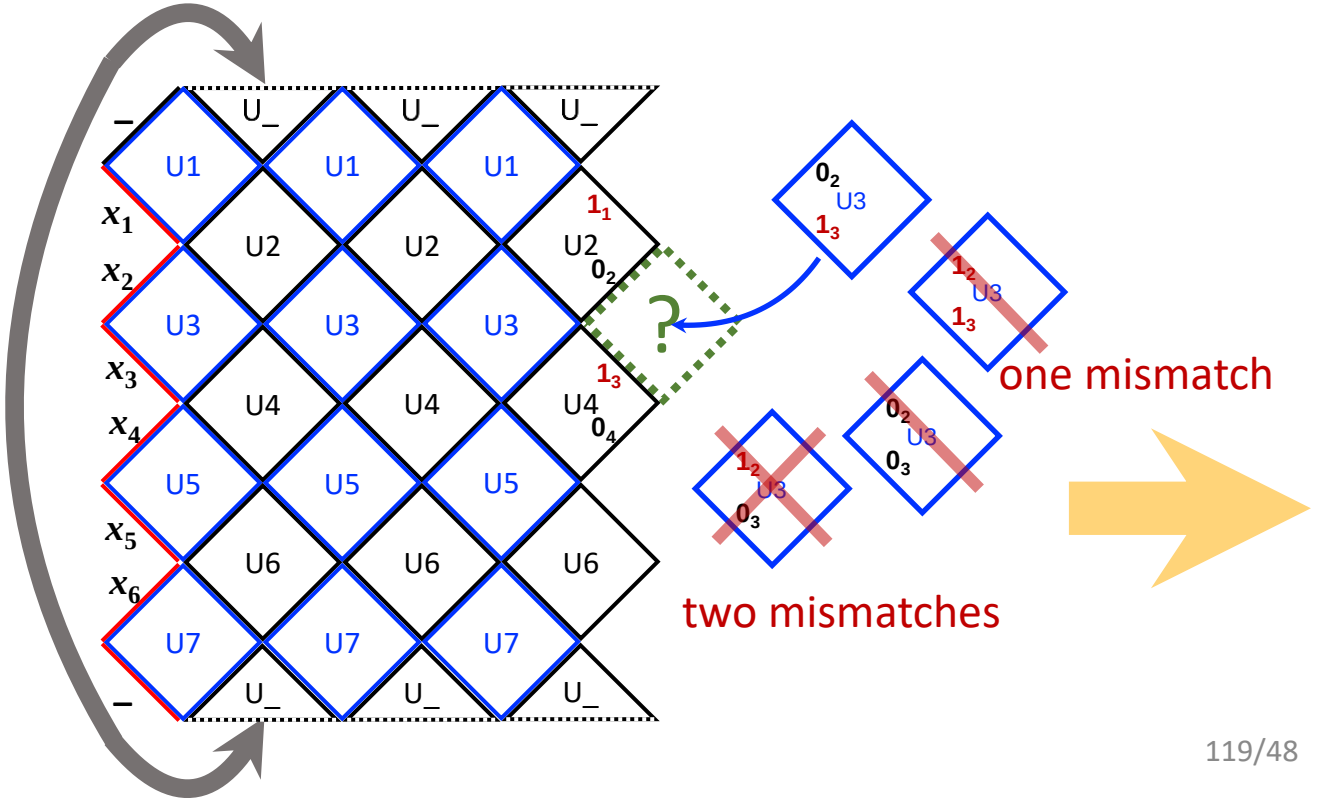

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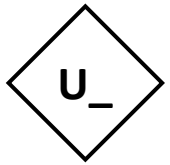

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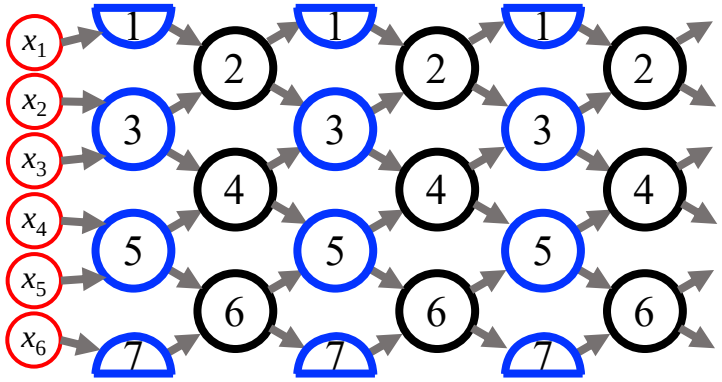

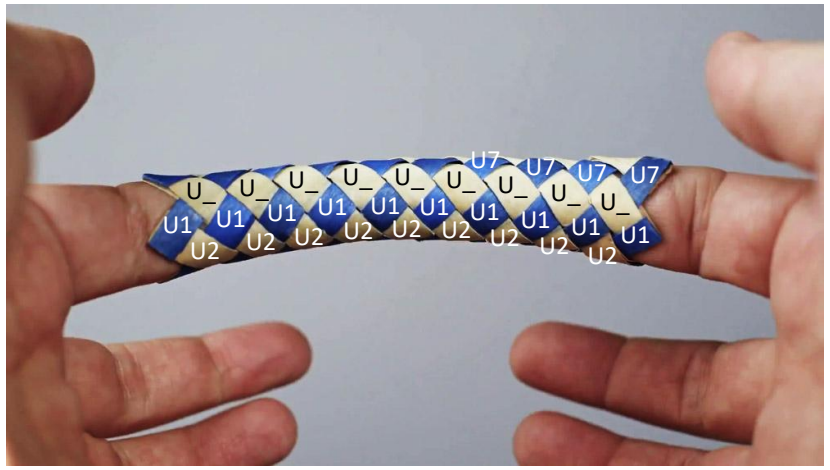

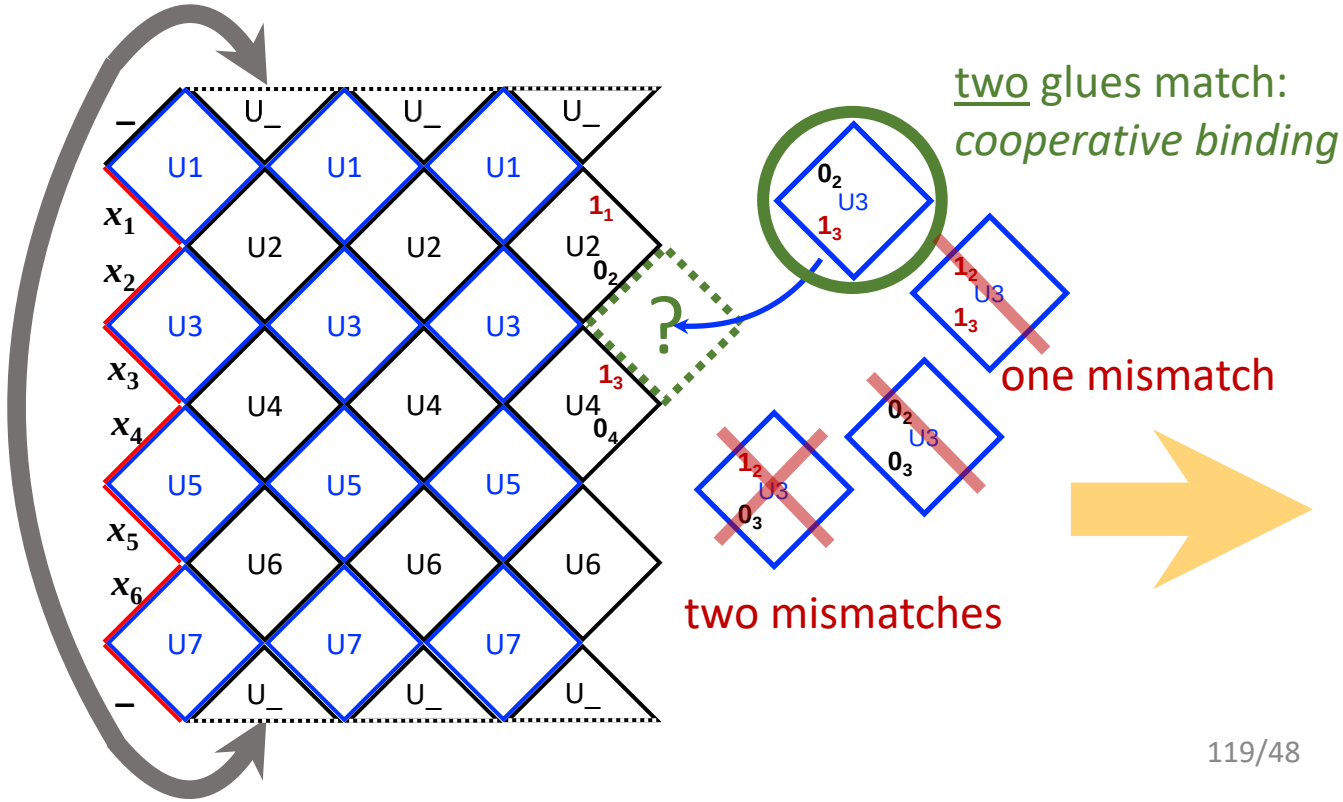

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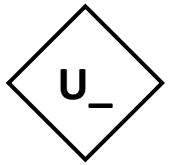

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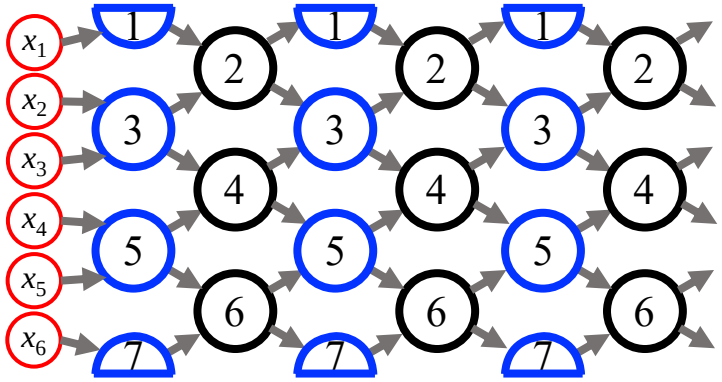
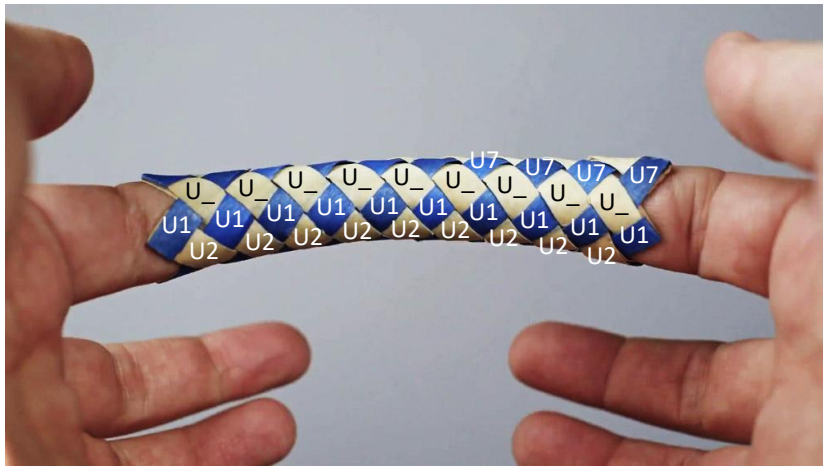
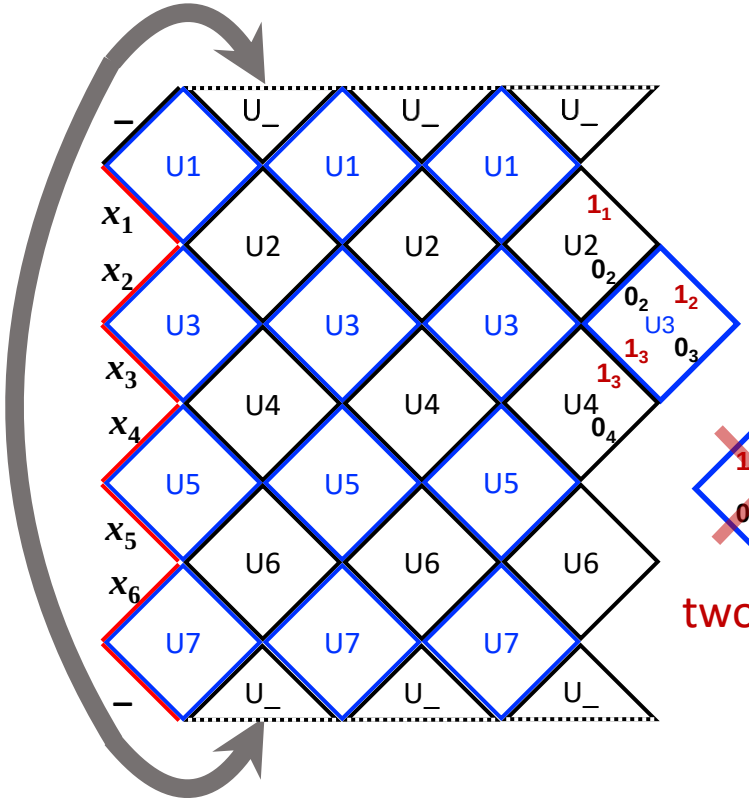
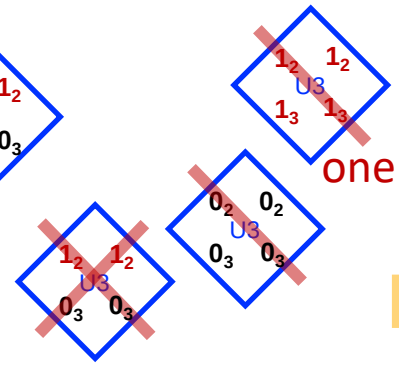






two glues match:
cooperative binding



one mismatch

two mismatches

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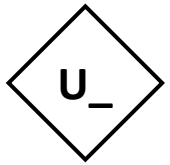


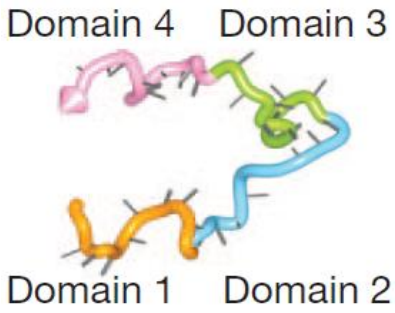
Hierarchy of abstractions

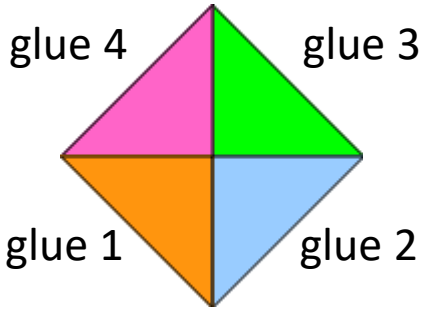
Bits: Boolean circuits compute

Tiles: Tile growth implements circuits

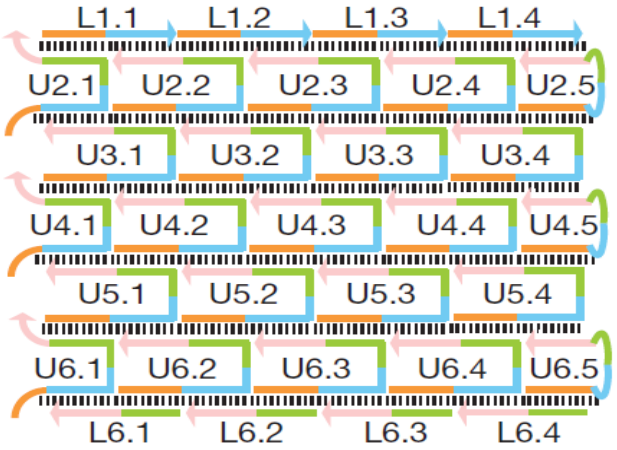
➡ **DNA: DNA strands implement tiles**

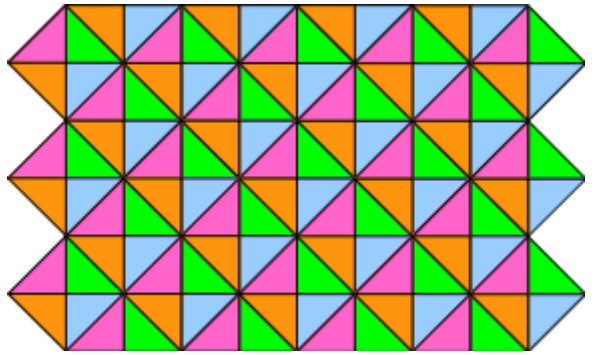
DNA single-stranded tiles





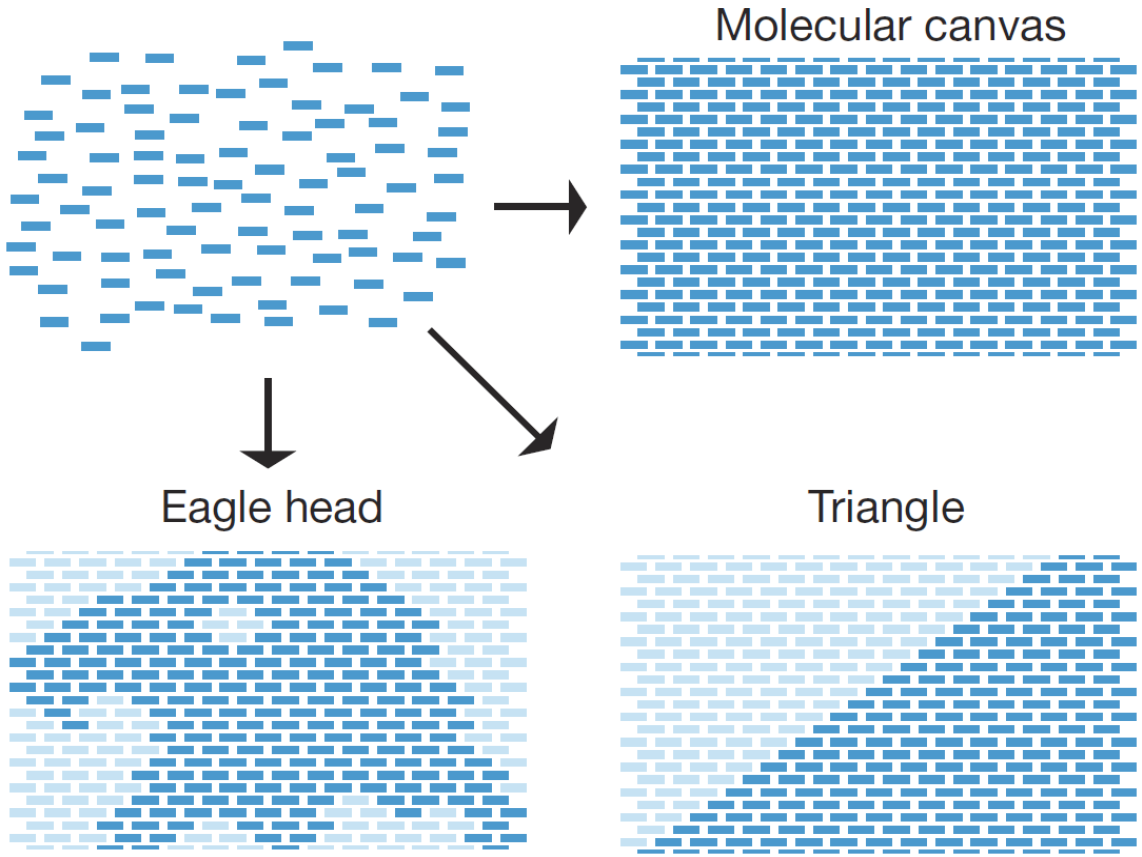
assembly





Yin, Hariadi, Sahu, Choi, Park, LaBean, and Reif.
Programming DNA tube circumferences.
Science 2008

Single-stranded tiles for making any shape



Bryan Wei, Mingjie Dai, and Peng Yin.
Complex shapes self-assembled from single-stranded DNA tiles.
Nature 2012.

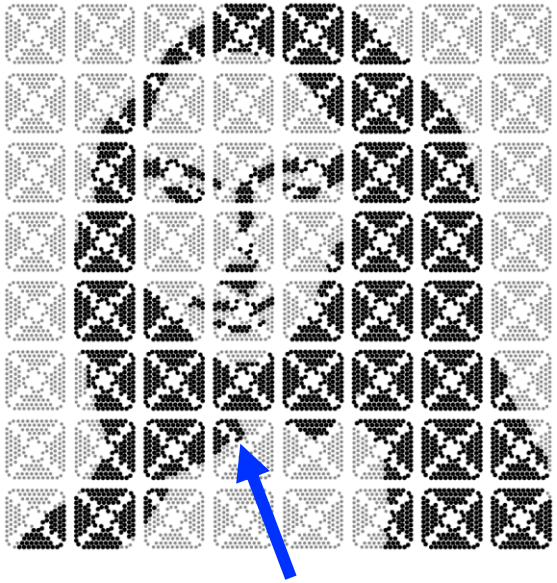
Uniquely addressed self-assembly versus algorithmic

Unique addressing: each DNA "monomer" appears **exactly once** in final structure.

single DNA origami

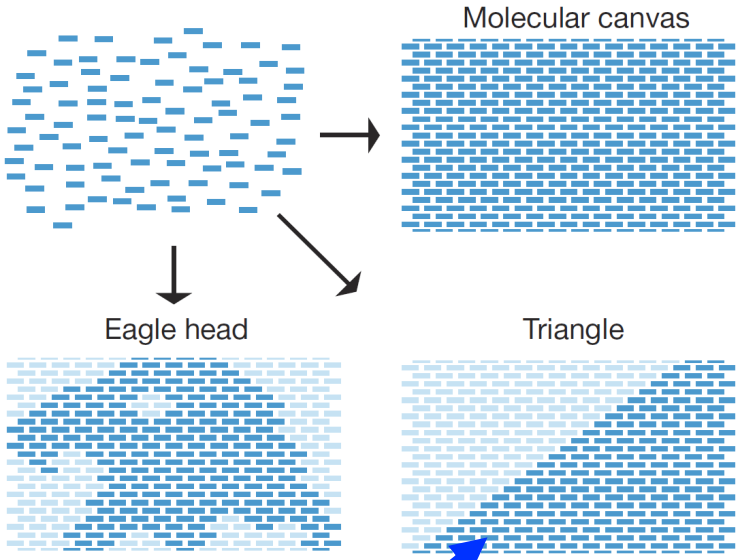
staple strand for position (4,2)

array of many DNA origamis



origami for position (4,2)

uniquely-addressed tiles



tile for position (4,2)

Uniquely addressed self-assembly versus algorithmic

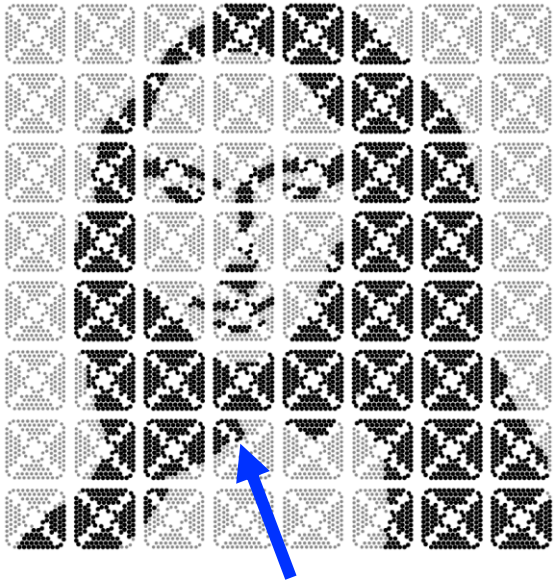
Unique addressing: each DNA "monomer" appears **exactly once** in final structure.

Algorithmic: DNA tiles are **reused** throughout the structure.

single DNA origami

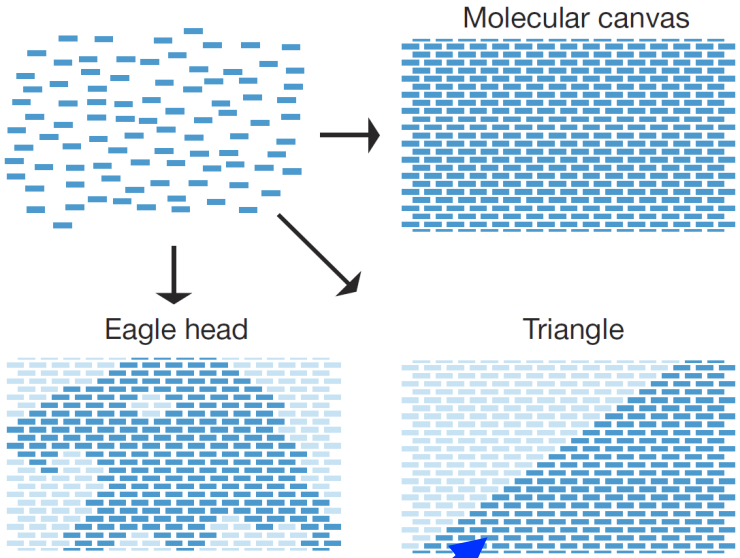
staple strand for position (4,2)

array of many DNA origamis



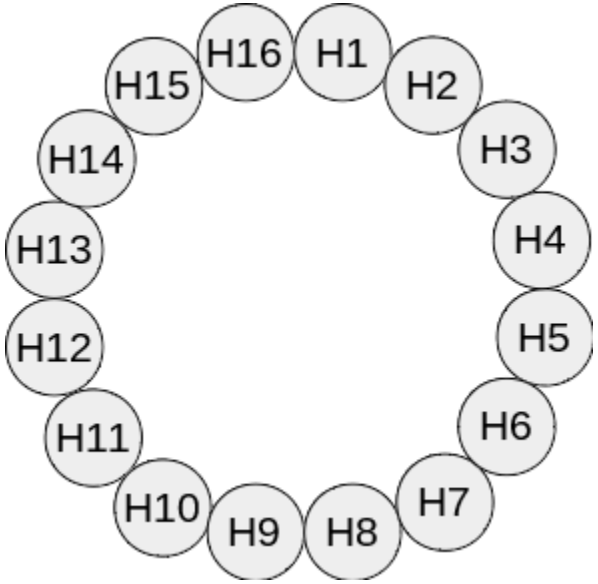
origami for position (4,2)

uniquely-addressed tiles

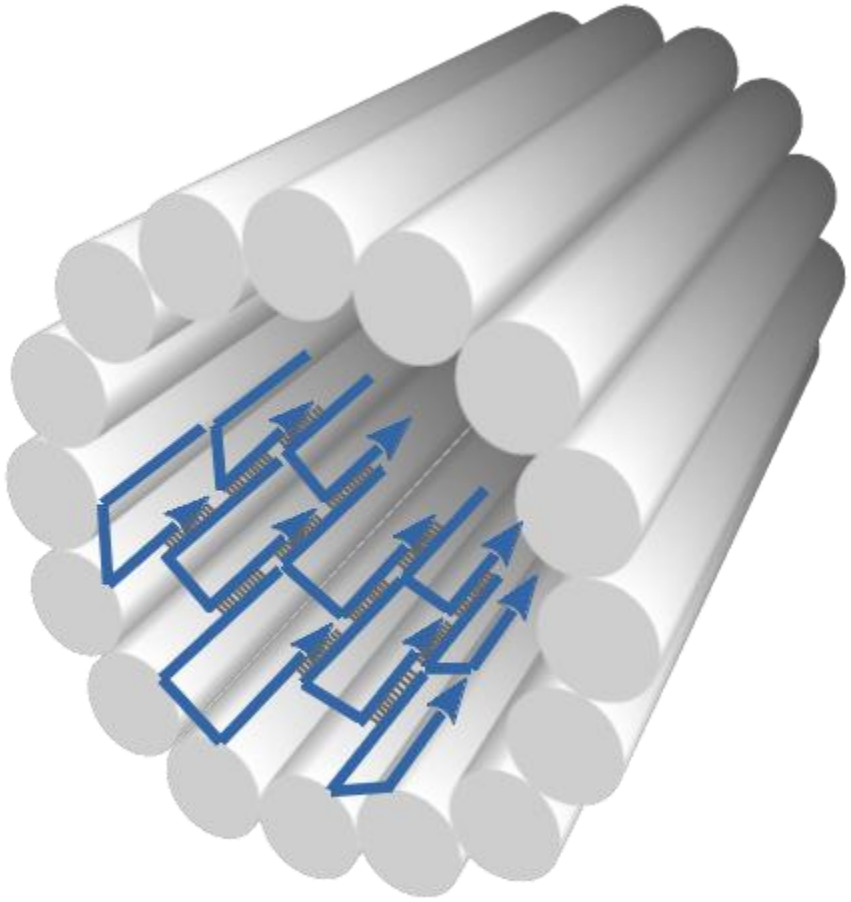


tile for position (4,2)

Single-stranded tile tubes

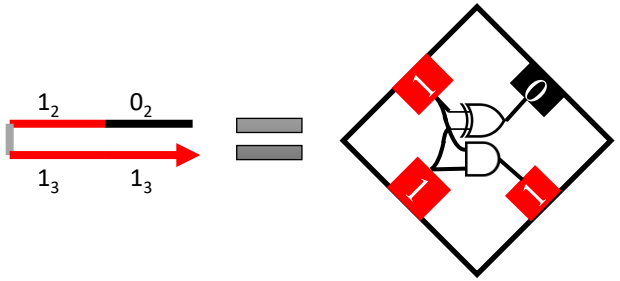


DNA-level diagram of 20-helix tube

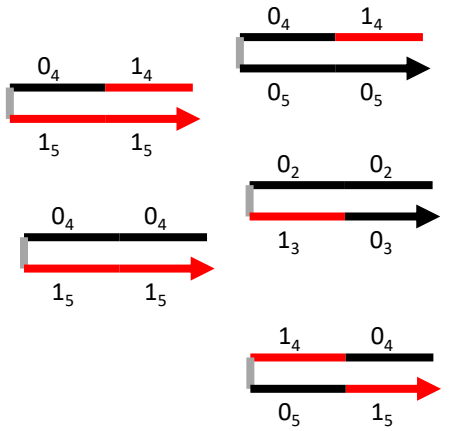


Yin, Hariadi, Sahu, Choi, Park, LaBean, and Reif. *Programming DNA tube circumferences*, Science 2008.

Seeded growth

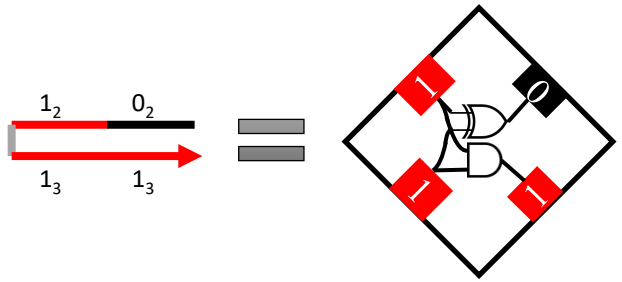


single-stranded tiles
implementing circuit gates



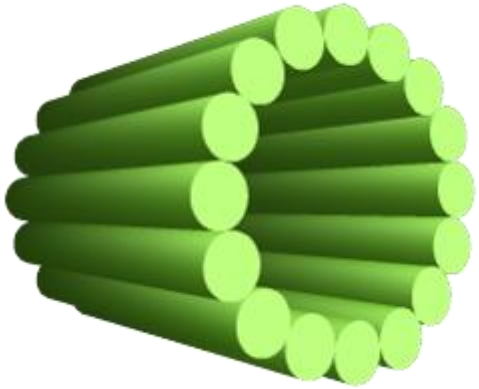
need barrier to <u>nucleation</u>
(tile growth without seed);
[tile]=100 nM;
temperature=50.9° C

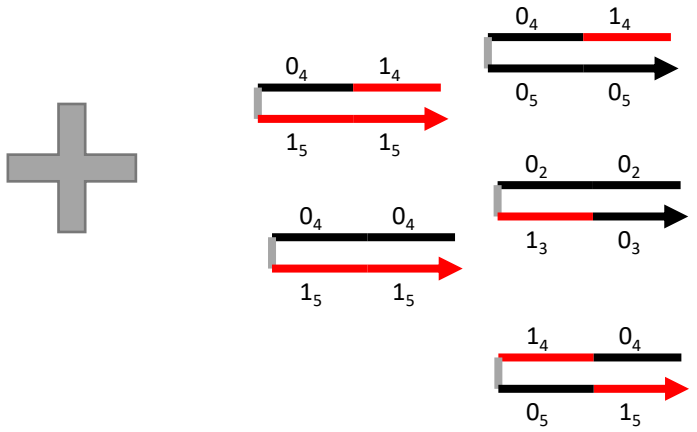
Seeded growth



DNA origami seed

single-stranded tiles
implementing circuit gates





need barrier to <u>nucleation</u>
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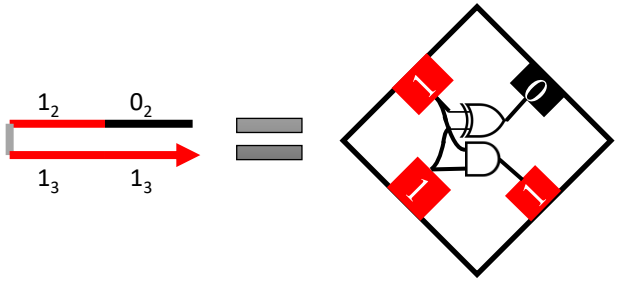
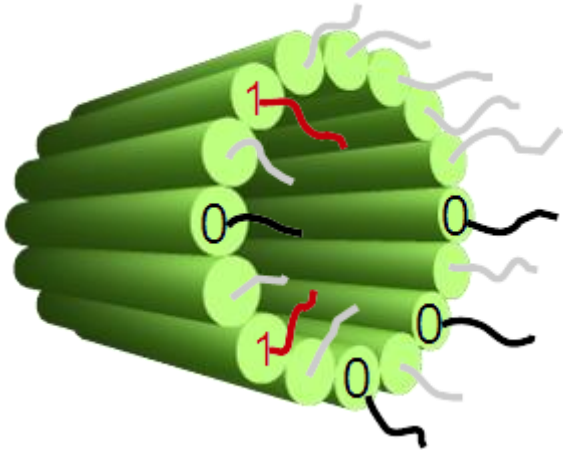
Seeded growth

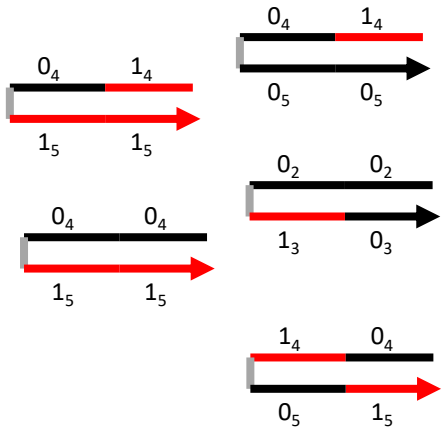


DNA origami seed

single-stranded “input-adapter” extensions encoding 6 input bits



single-stranded tiles implementing circuit gates



need barrier to <u>nucleation</u>
(tile growth without seed);
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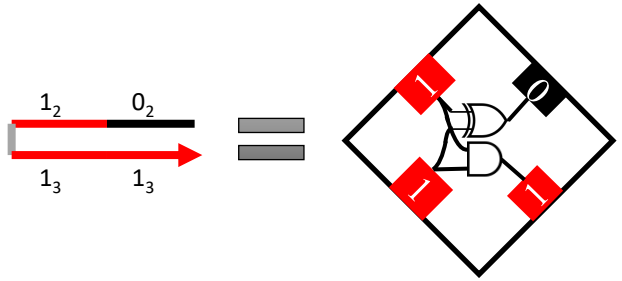
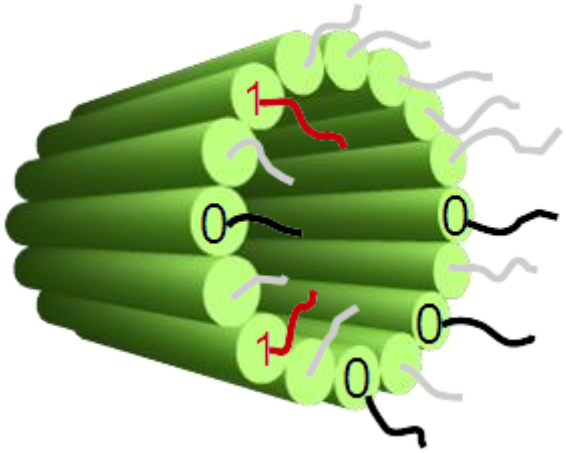
temperature=50.9° C

Seeded growth

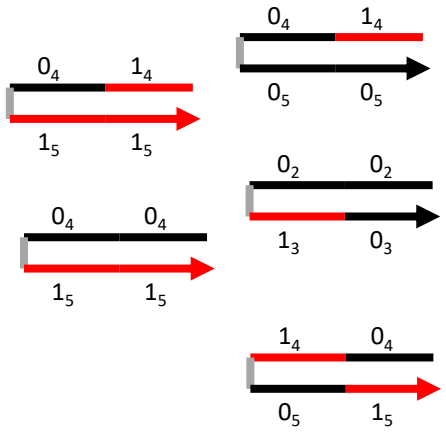


DNA origami seed

single-stranded “input-adapter” extensions encoding 6 input bits

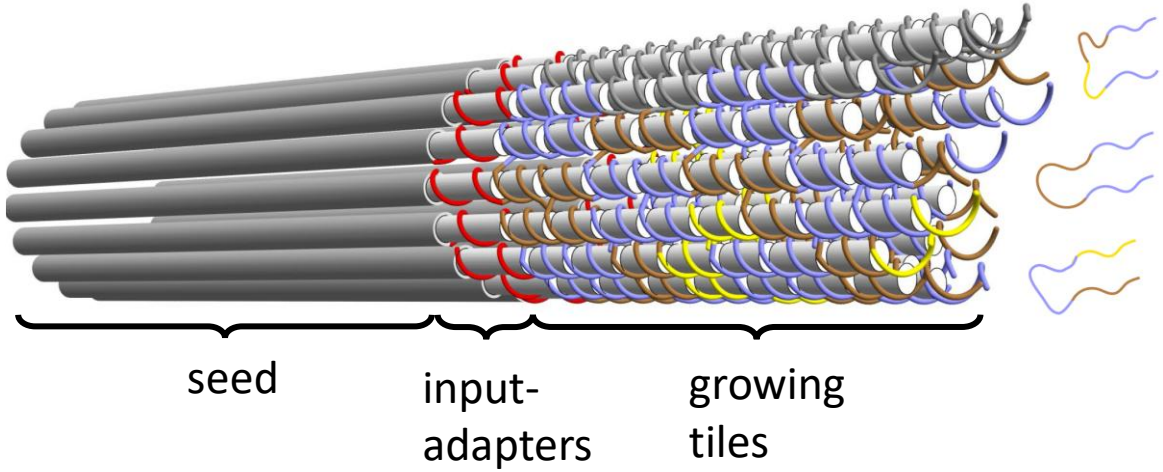


single-stranded tiles implementing circuit gates

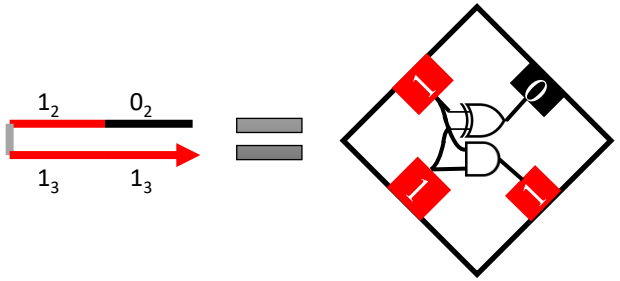


need barrier to <u>nucleation</u> (tile growth without seed);
[tile]=100 nM;
temperature=50.9° C

hold 8-48 hours

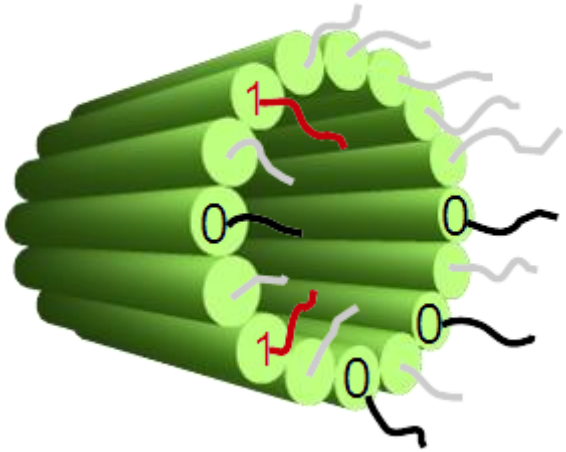


Seeded growth

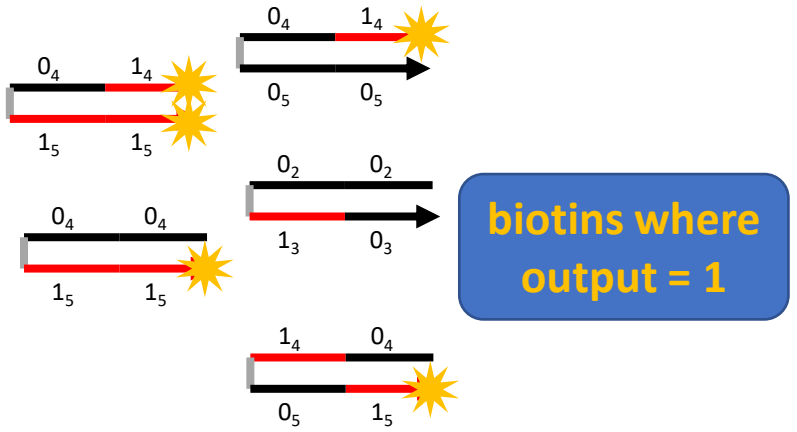


DNA origami seed

single-stranded “input-adapter” extensions encoding 6 input bits



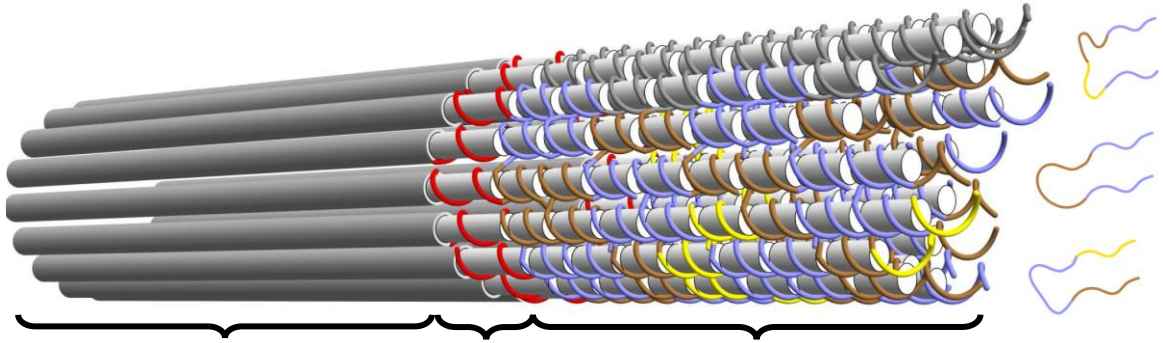
single-stranded tiles implementing circuit gates



biotins where output = 1

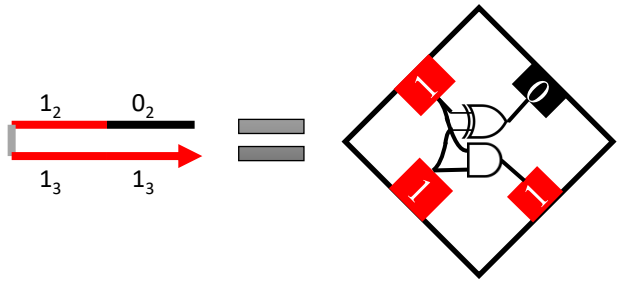
need barrier to nucleation (tile growth without seed); [tile]=100 nM; temperature=50.9° C

hold 8-48 hours

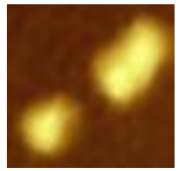


seed | input-adapters | growing tiles

Seeded growth

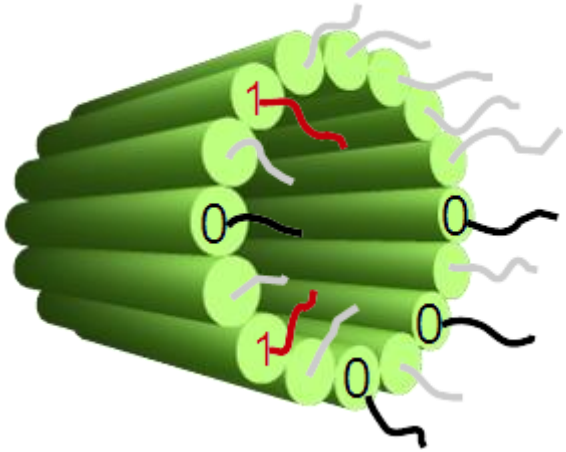


can later add streptavidin (5 nm wide protein) to bind biotins and visualize where the 1's are

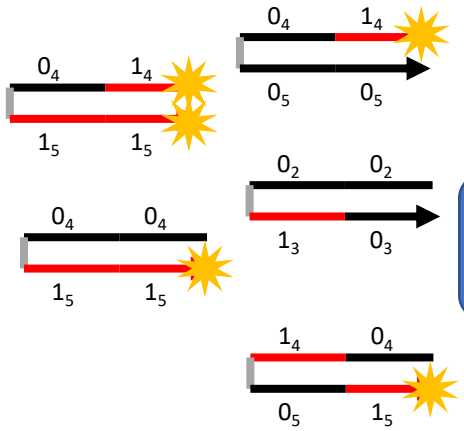


DNA origami seed

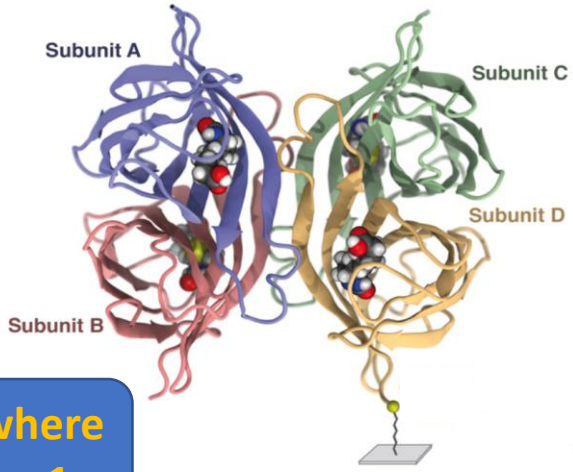
single-stranded "input-adapter" extensions encoding 6 input bits



single-stranded tiles implementing circuit gates

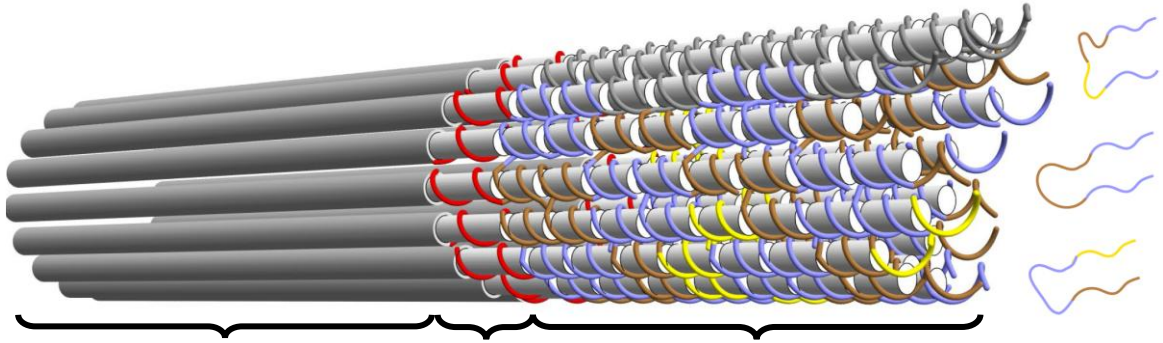


biotins where output = 1



need barrier to nucleation (tile growth without seed);
[tile]=100 nM;
temperature=50.9° C

hold 8-48 hours



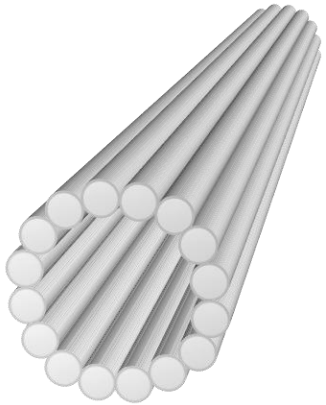
seed

input-adapters

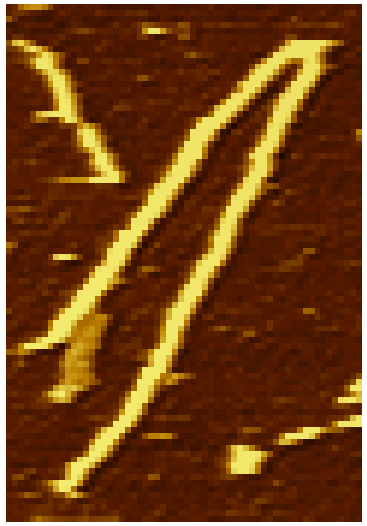
growing tiles

Tubes to ribbons

tube



AFM image

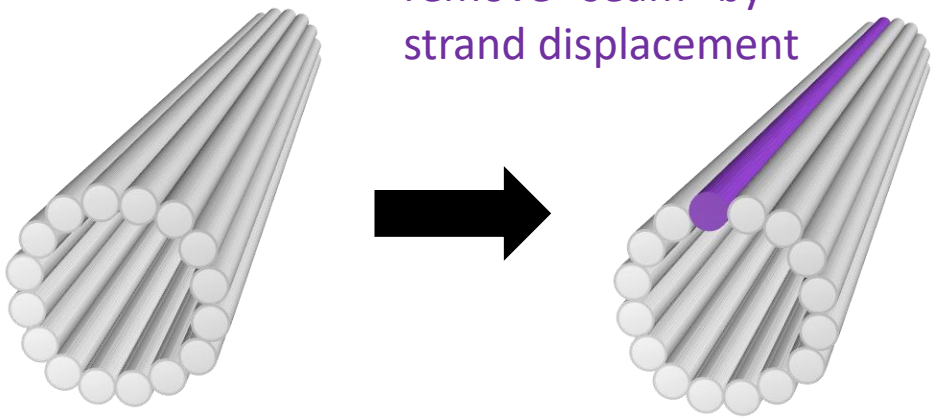


500 nm

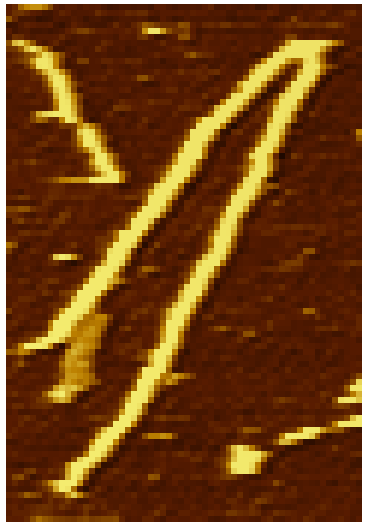
Tubes to ribbons

tube

remove “seam” by
strand displacement



AFM
image



500 nm

Tubes to ribbons

tube

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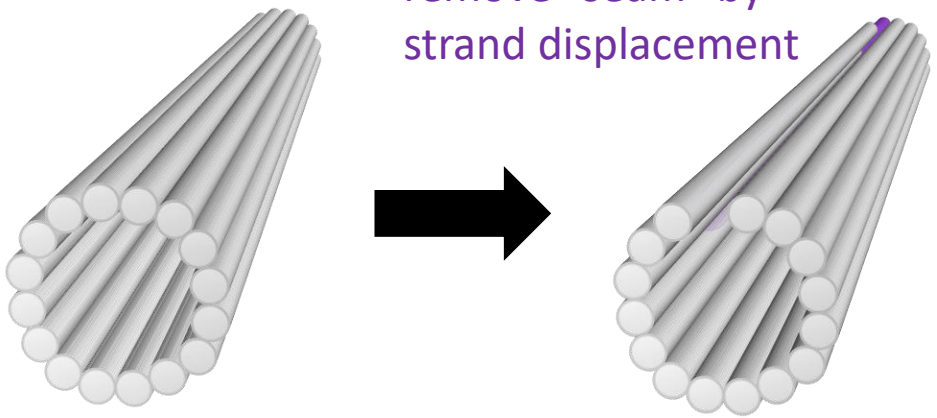

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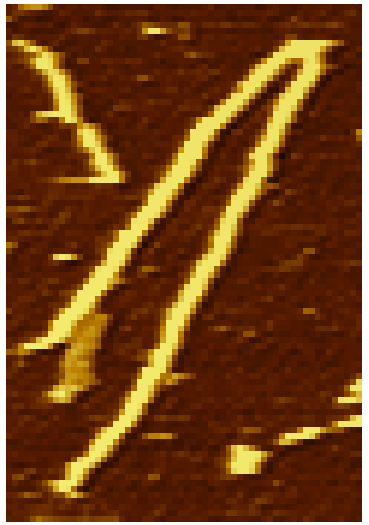
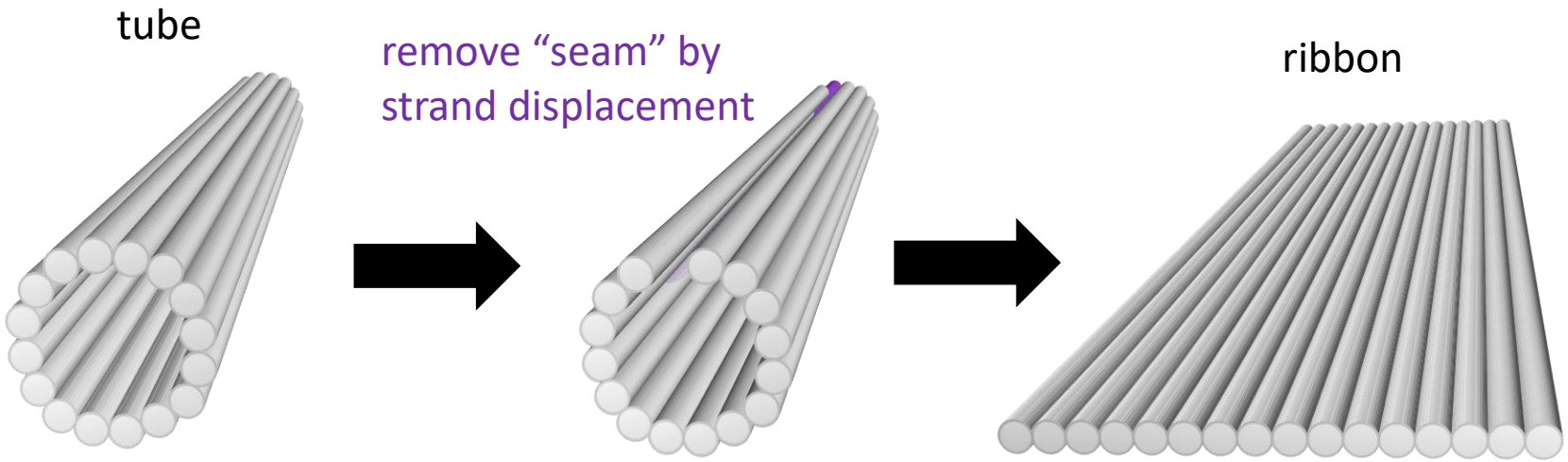
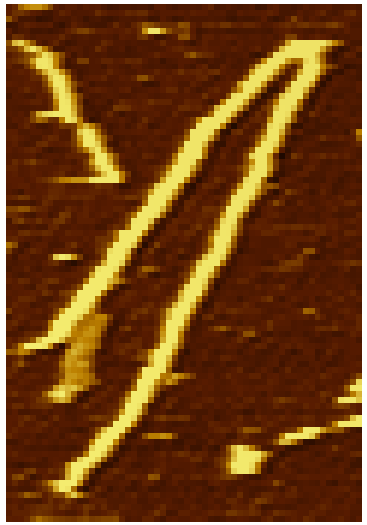


500 nm

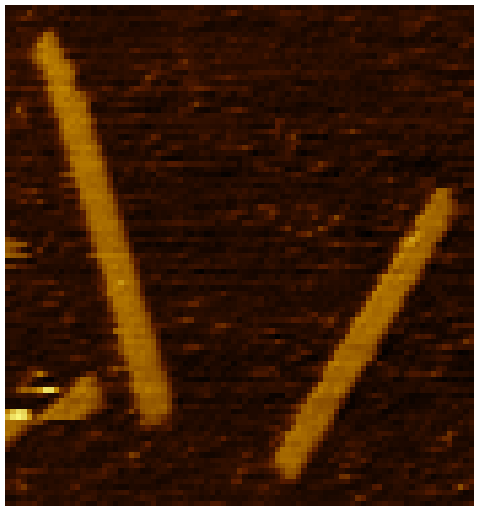
Tubes to ribbons



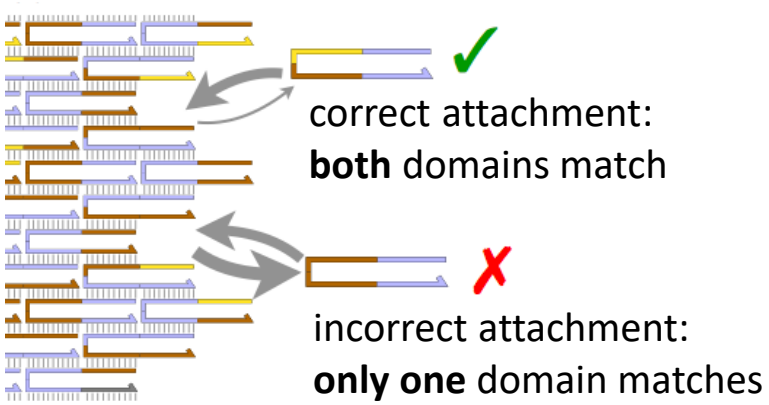
AFM
image



500 nm

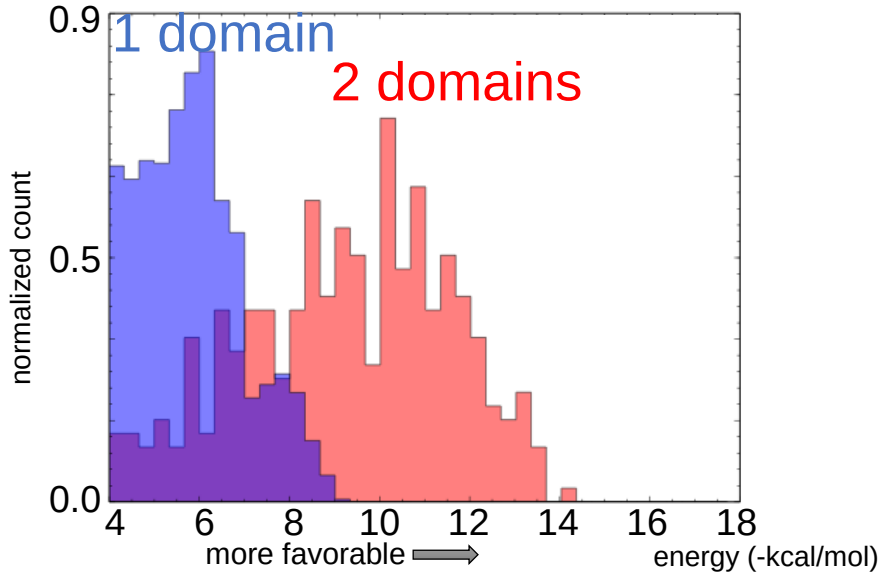


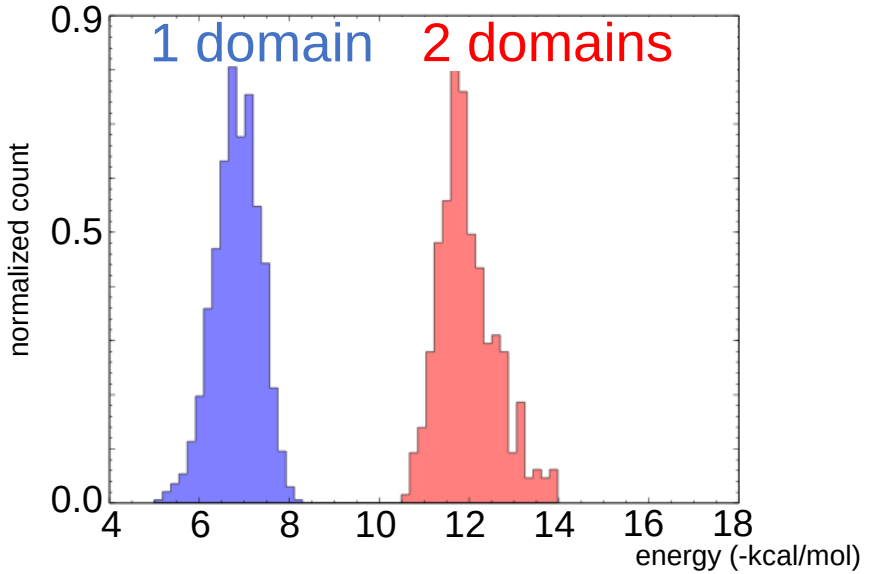
DNA sequence design

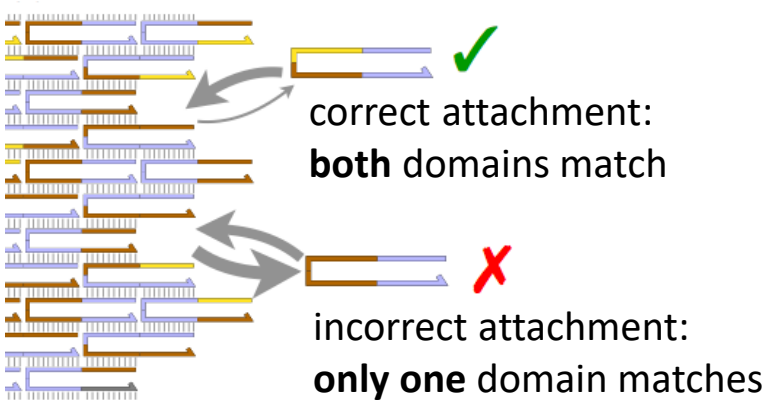


DNA sequence design

Random sequences vs designed sequences





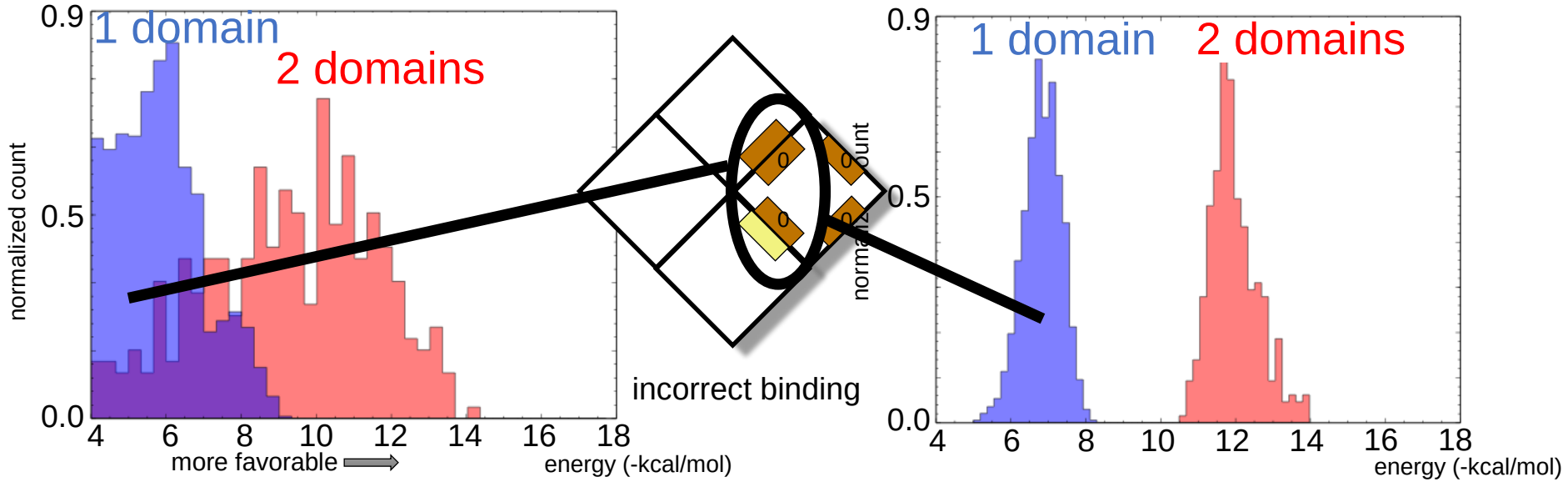


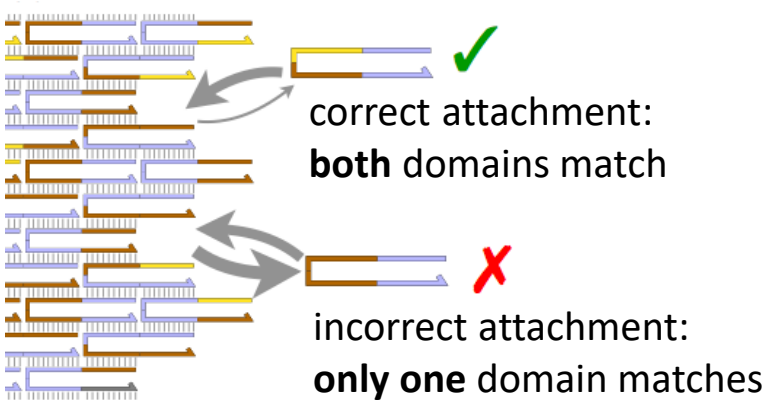
✓ correct attachment:
both domains match

✗ incorrect attachment:
only one domain matches

DNA sequence design

Random sequences vs designed sequences

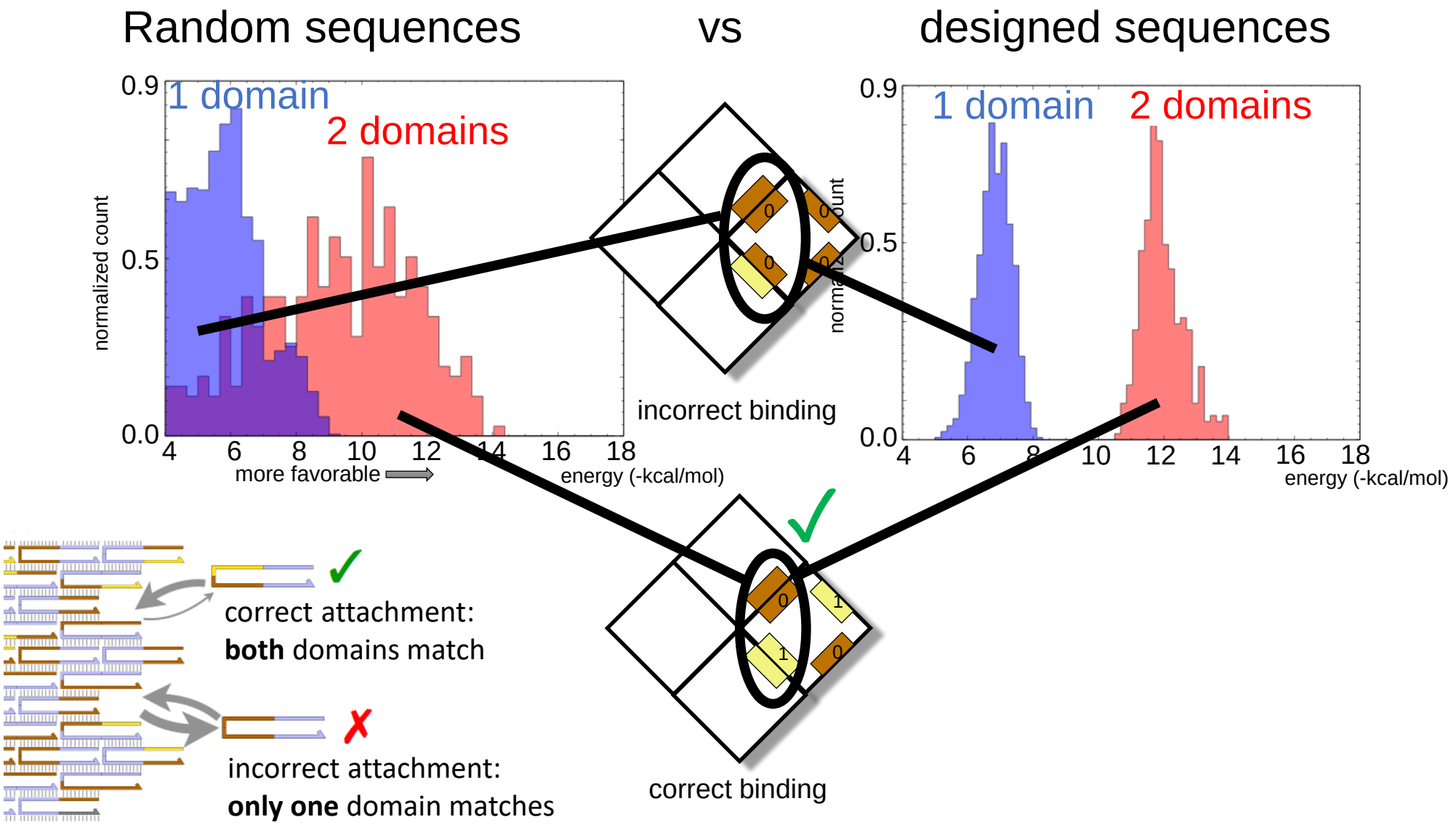




correct attachment:
both domains match

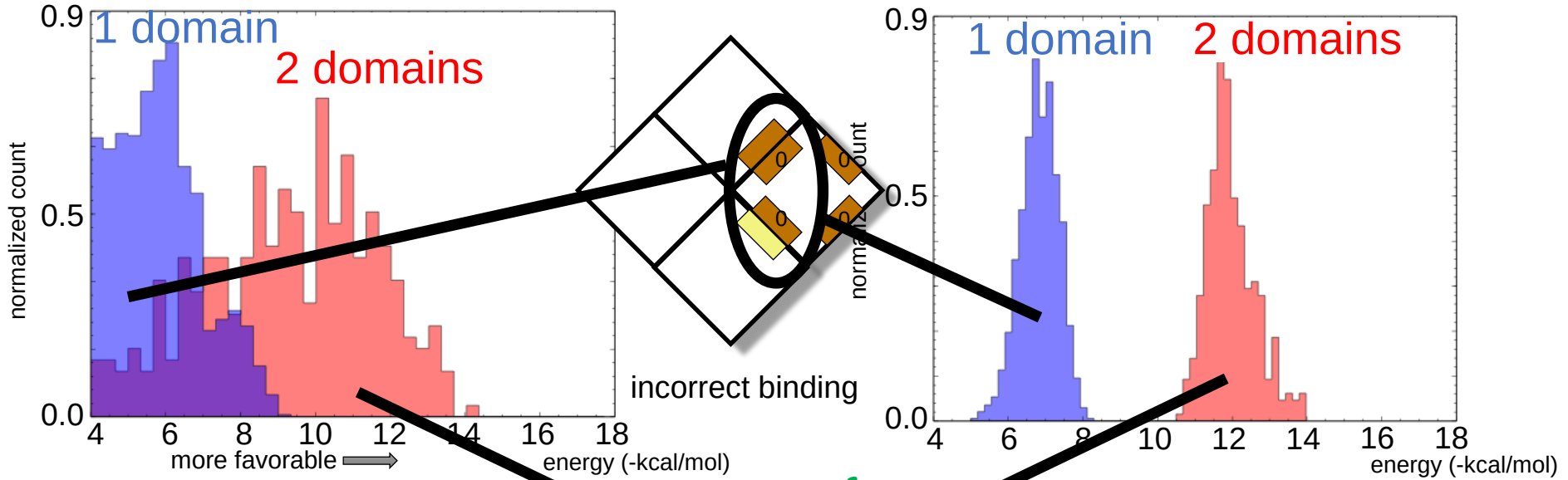
incorrect attachment:
only one domain matches

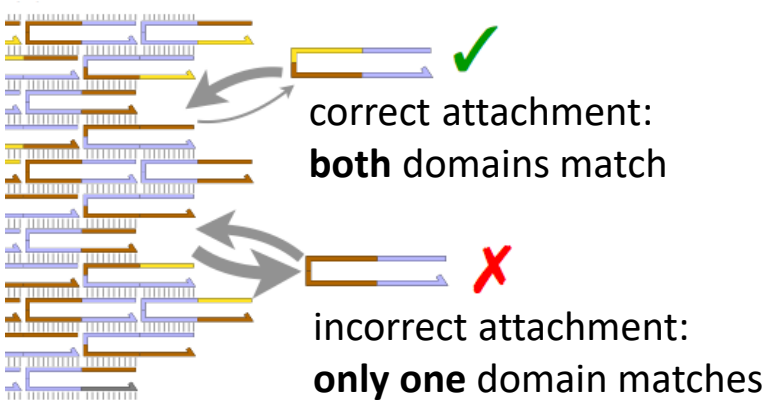
DNA sequence design



DNA sequence design

Random sequences vs designed sequences





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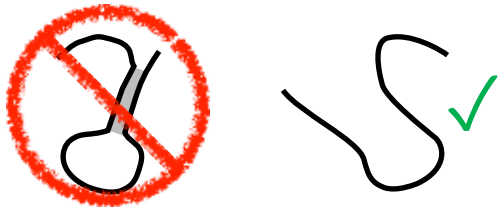
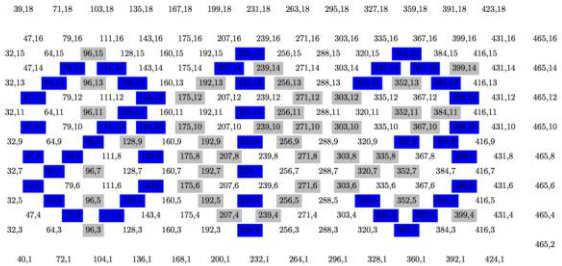


Other goals:

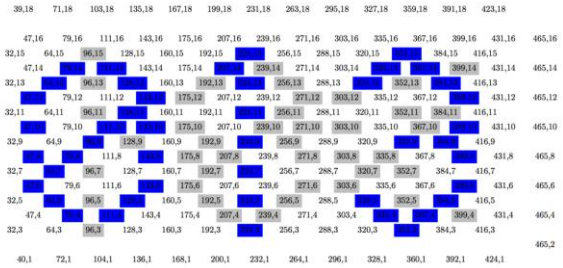
- low strand secondary structure
- low interaction between strands

Bar-coding origami seed for imaging multiple samples at once



some staples of origami seed have version with a biotin

Bar-coding origami seed for imaging multiple samples at once

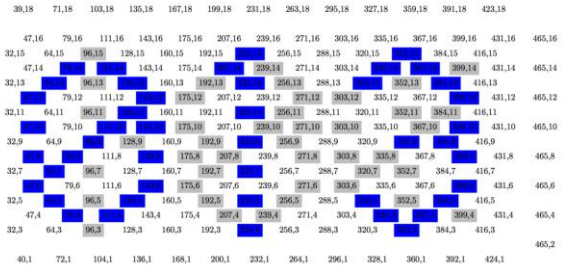


some staples of origami seed have version with a biotin

Generate plate map

```
*********************************************
* pos3=u, coding staples, no biotin (14 staples)
16H barrel stap 2
  1  2  3  4  5  6  7  8  9 10 11 12 |
A                                    |
B                       X  X         |
C                   X                |
D                      X     X       |
E                                X   |
F                X  X     X     X    |
G                      X  X  X       |
H                                    |
pos3=u, coding staples, biotin (16 staples)
16H 3bit biotin staples              | unzippers2 + biotin_staples_2_3      |
  1  2  3  4  5  6  7  8  9 10 11 12 |  1  2  3  4  5  6  7  8  9 10 11 12 |
A                       X  X     X   | A                                    |
B                       X     X  X   | B                                    |
C                       X     X  X   | C                                    |
D                             X  X   | D                                    |
E                       X     X      | E                  X                 |
F                                    | F                  X                 |
G                                    | G                                    |
H             X                      | H                                    |
*********************************************
```

Bar-coding origami seed for imaging multiple samples at once



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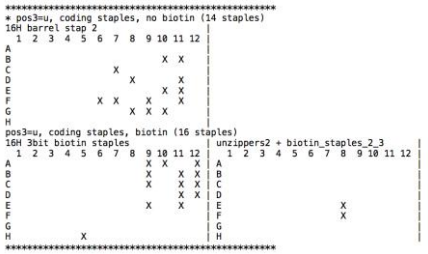
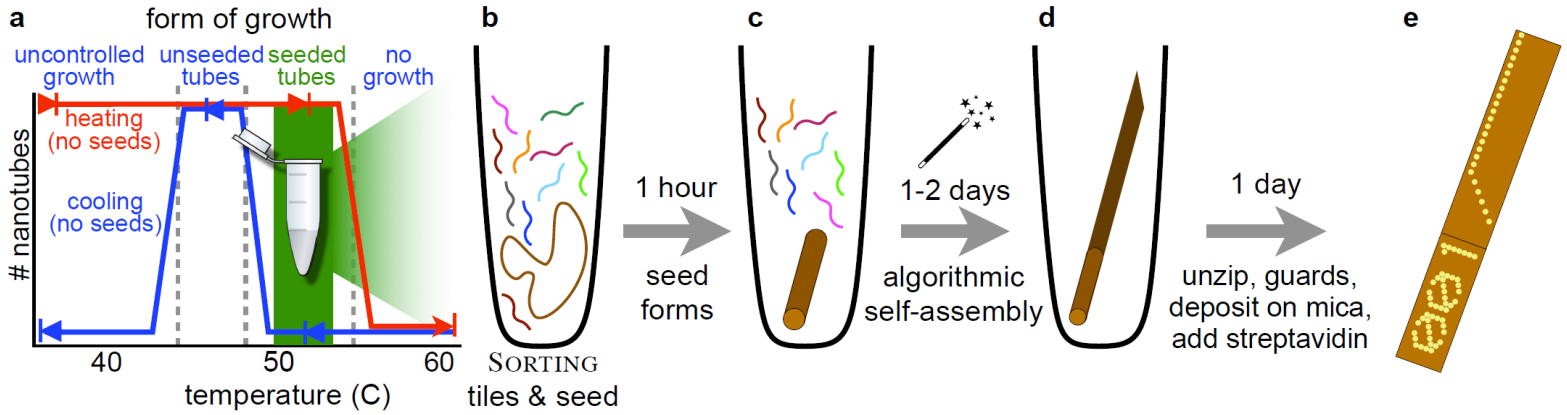
Generate plate map



represents some combination of circuit and input, e.g.,
013 = *“parity circuit, input=011010”*

label with streptavidin

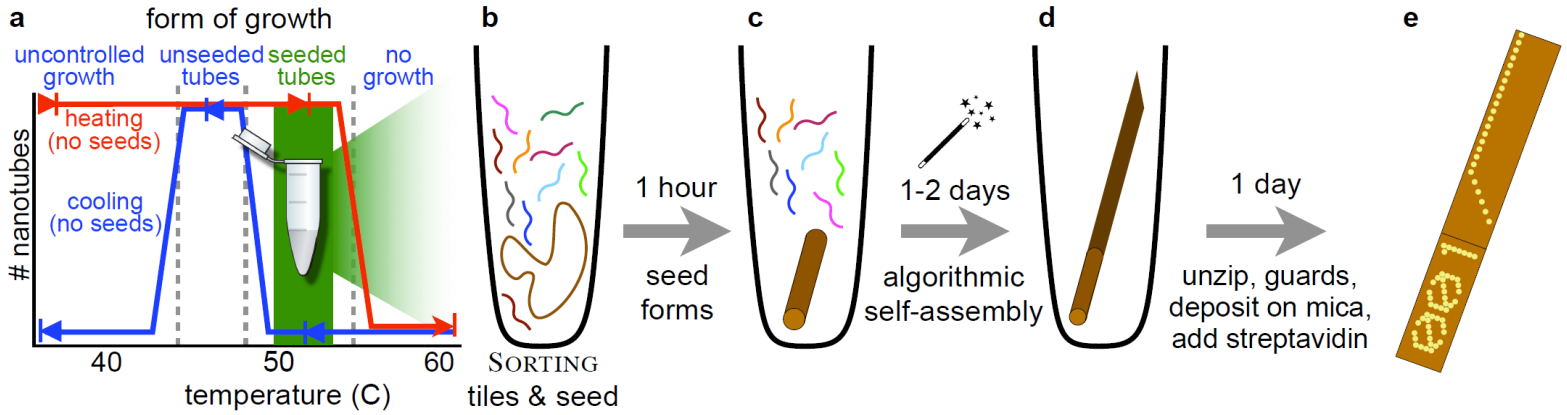
Experimental protocol



To execute circuit γ on input $x \in \{0,1\}^*$:

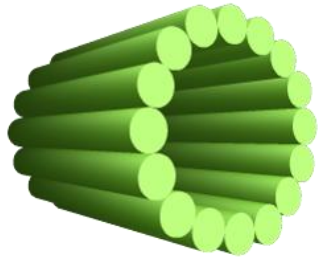
- Mix

Experimental protocol

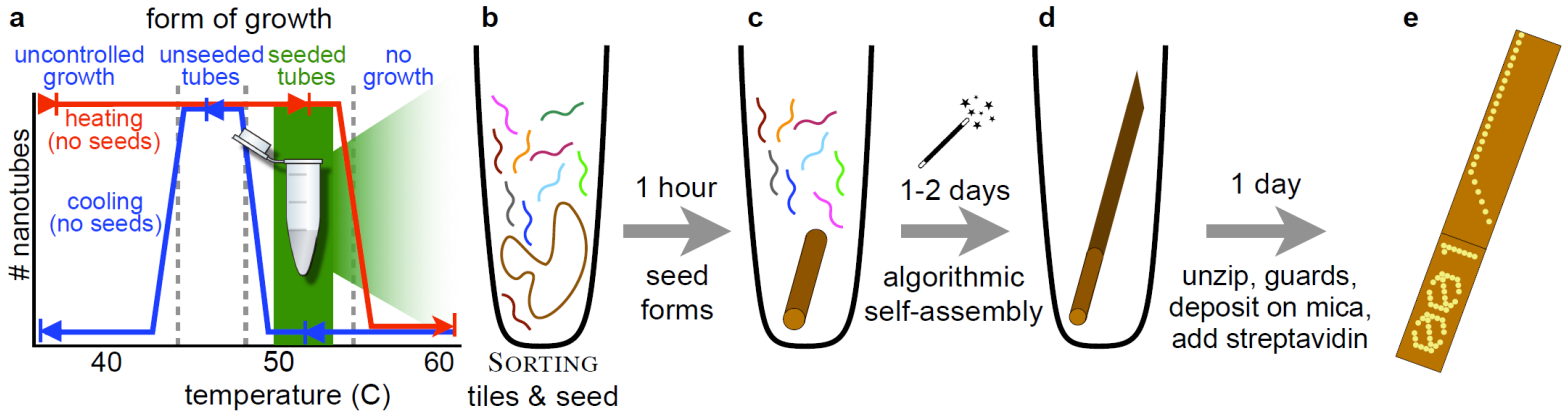


To execute circuit γ on input $x \in \{0,1\}^*$:

- Mix
  - origami seed (bar-coded to identify γ and x)

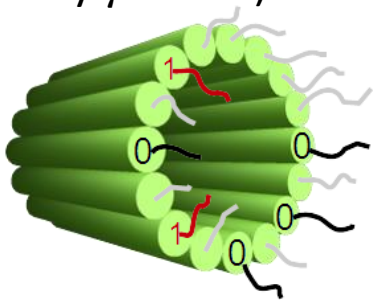


Experimental protocol

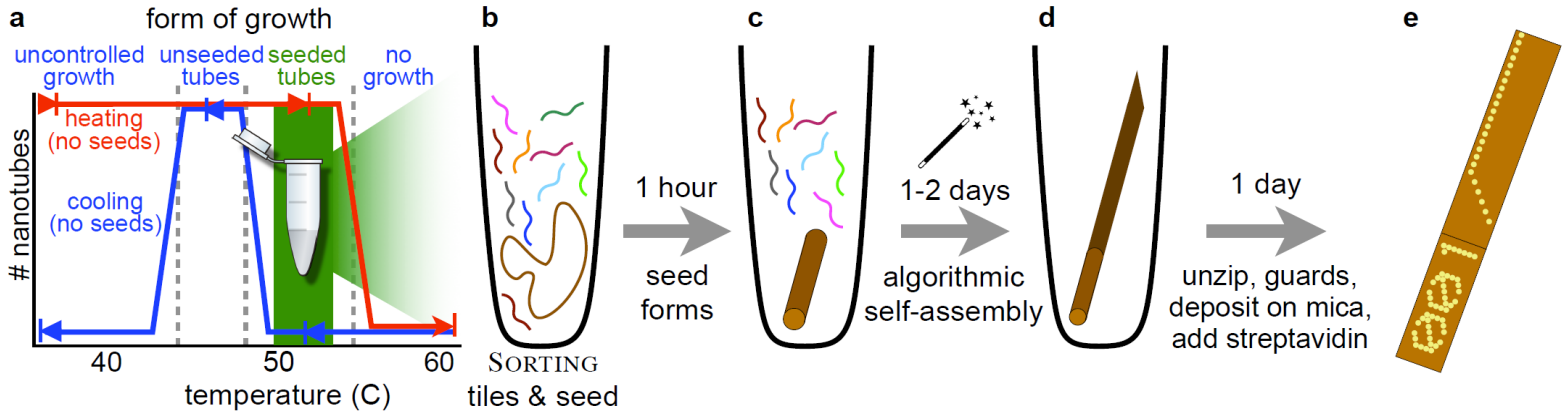


To execute circuit γ on input $x \in \{0,1\}^*$:

- Mix
  - origami seed (bar-coded to identify γ and x)
  - "adapter" strands encoding x



Experimental protocol



To execute circuit γ on input $x \in \{0,1\}^*$:

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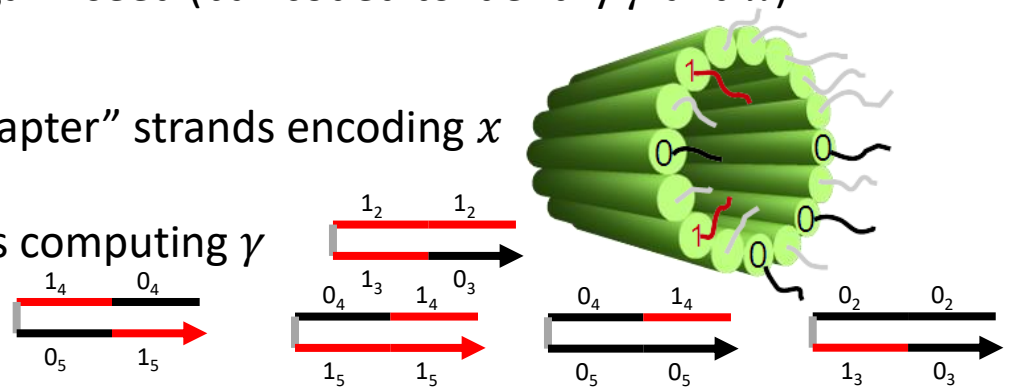
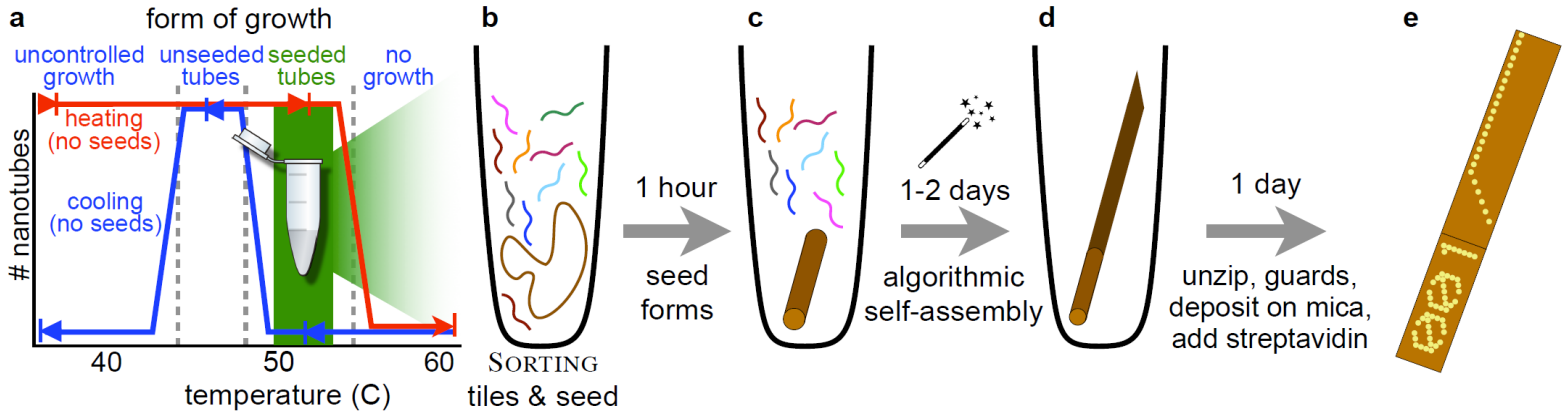
  - tiles computing γ



Experimental protocol



To execute circuit γ on input $x \in \{0,1\}^*$:

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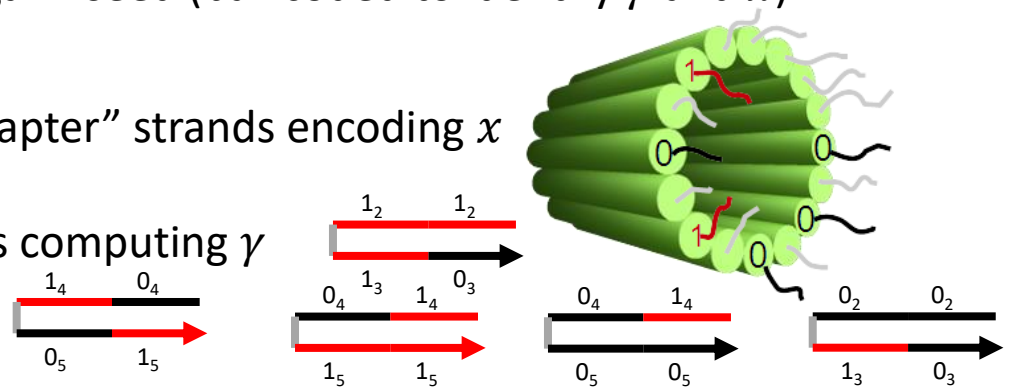
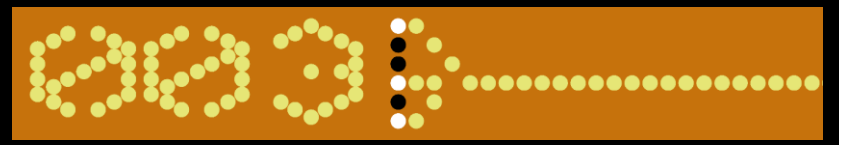


- Anneal 90° C to 50.9° C in 1 hour (*origami seeds form*)
- Hold at 50.9° C for 1-2 days (*tiles grow tubes from seed*)
- Add "unzipper" strands (remove seam to convert tube to ribbon)
- Add "guard" strands (complements of output sticky ends, to deactivate tiles)
- Deposit on mica, buffer wash, add streptavidin, AFM

Results

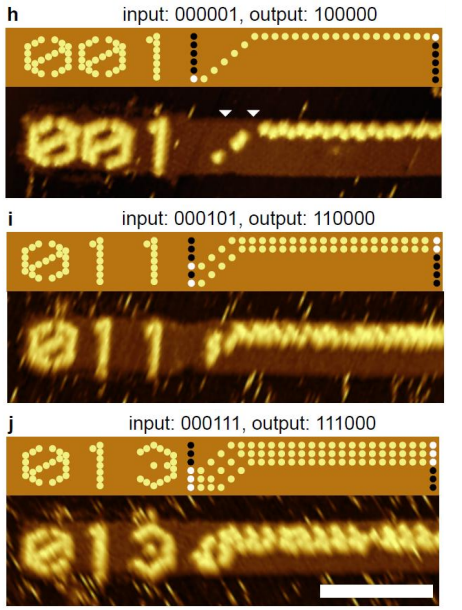
```
def test_parity():
    actual = parity('100101')

    expected =
```

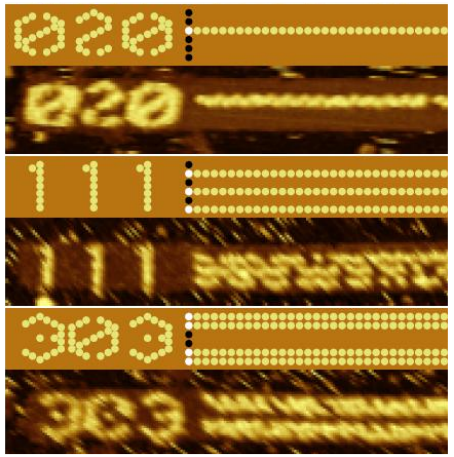


```
    assertEquals(expected, actual)
```

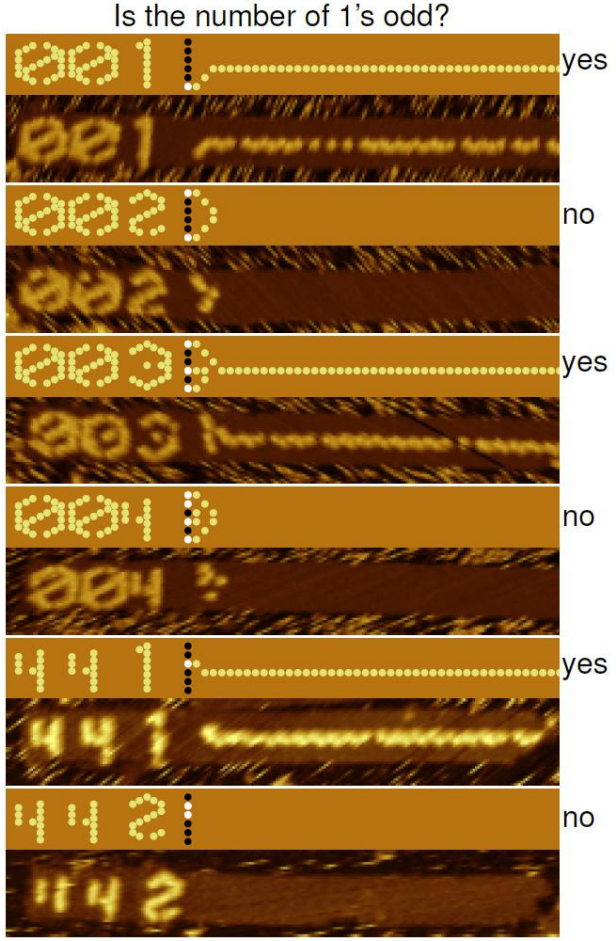
Sorting



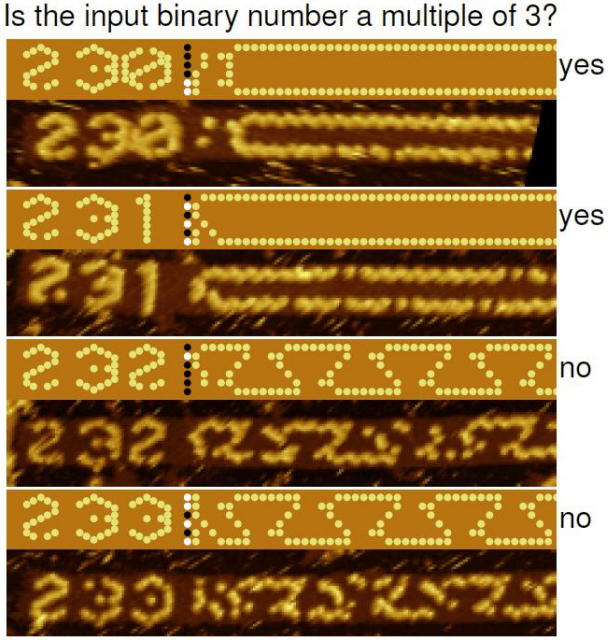
Copy



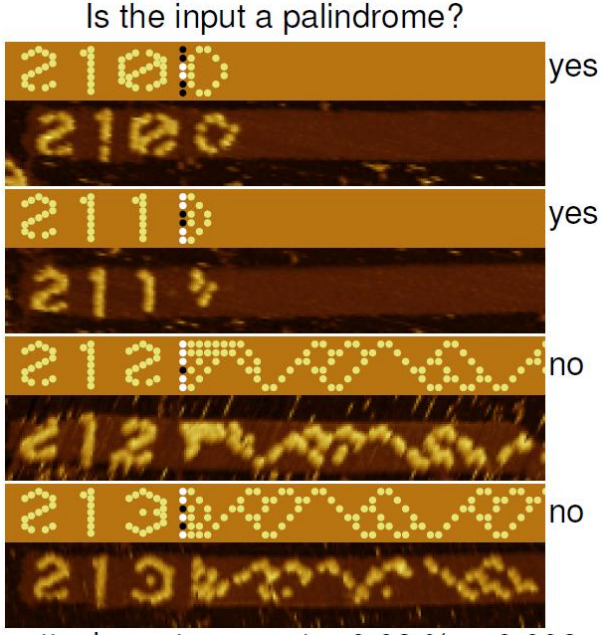
Parity



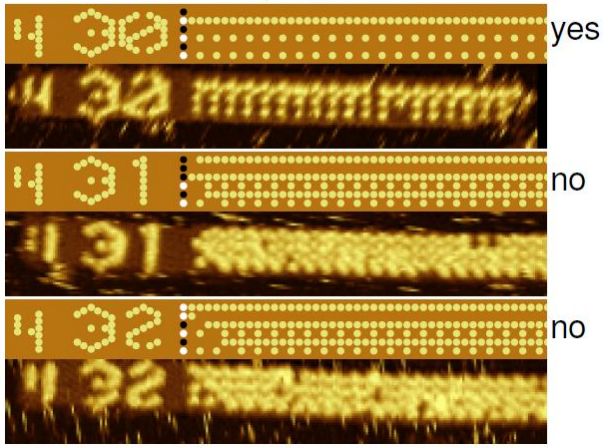
MultipleOf3



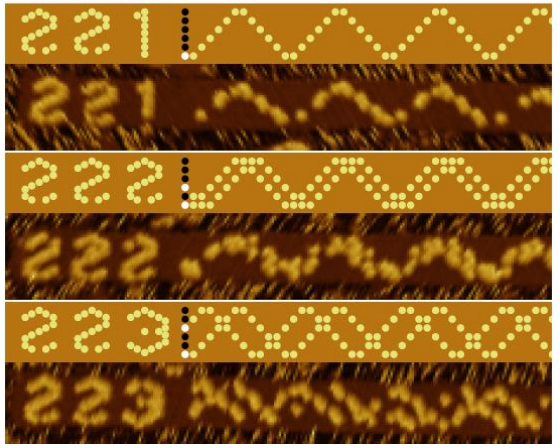
Palindrome



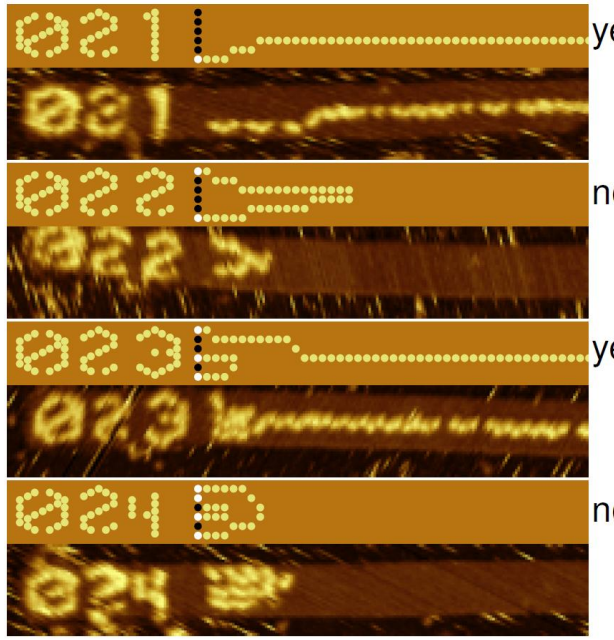
Recognise21



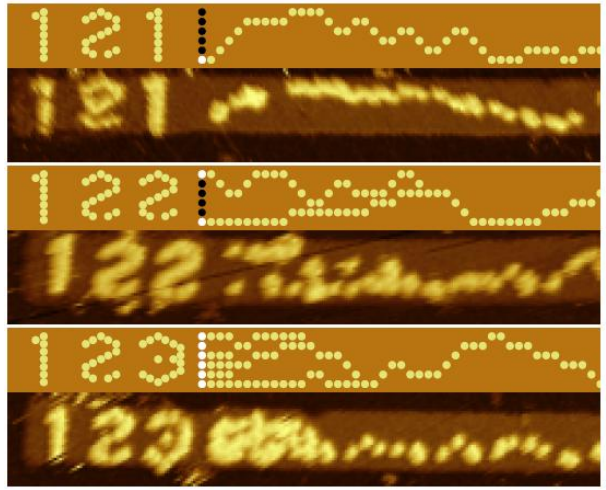
Zig-Zag



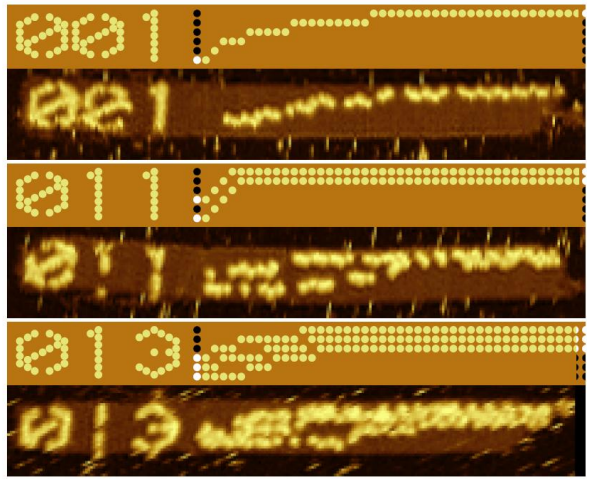
LazyParity



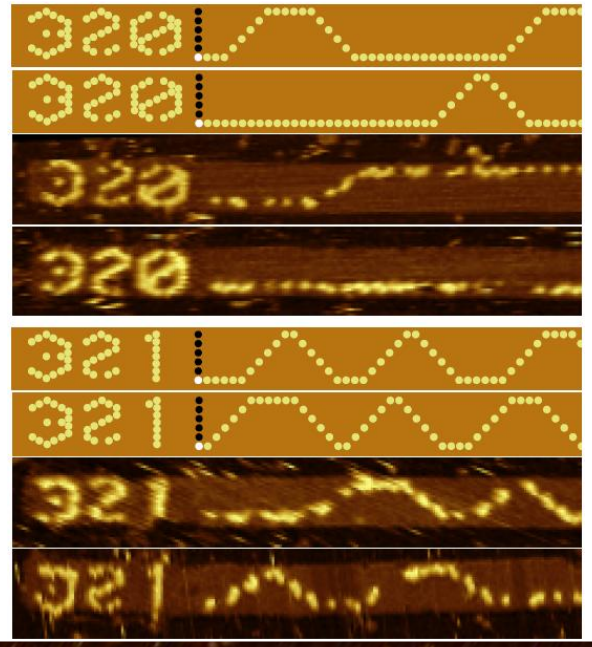
LeaderElection



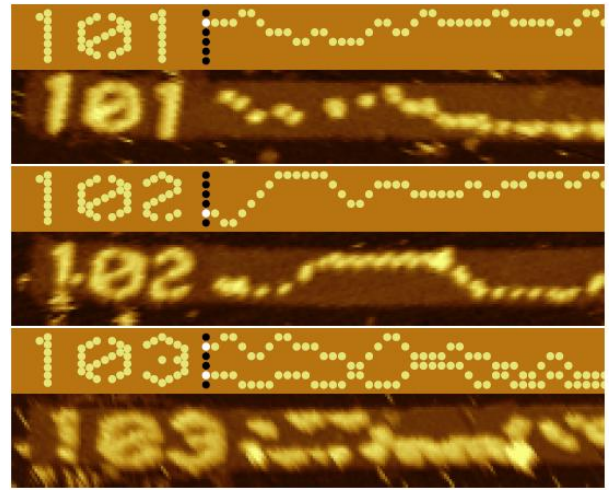
LazySorting



Waves

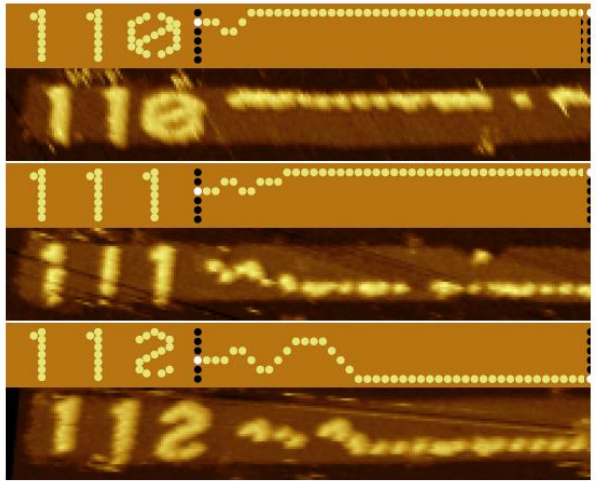


RandomWalkingBit

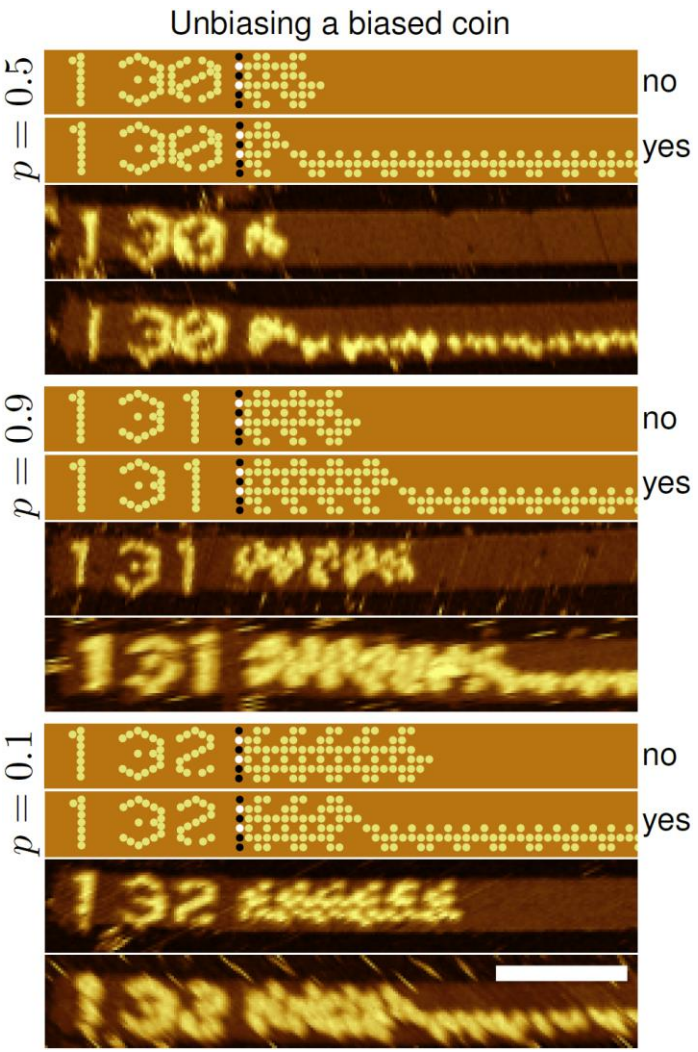


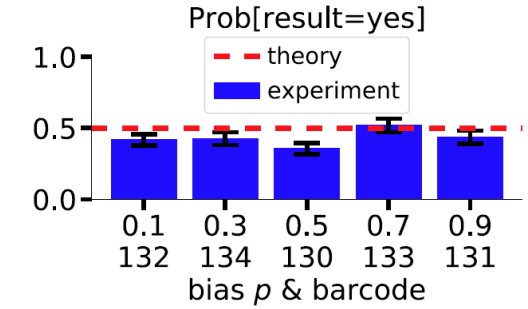
AbsorbingRandomWalkingBit

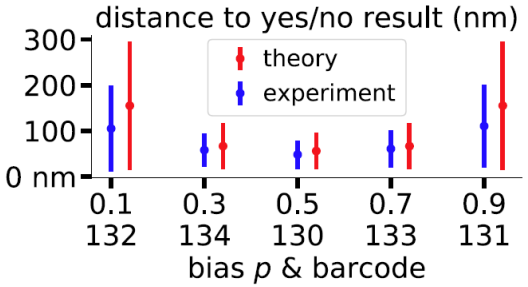
Random walker absorbs to top/bottom



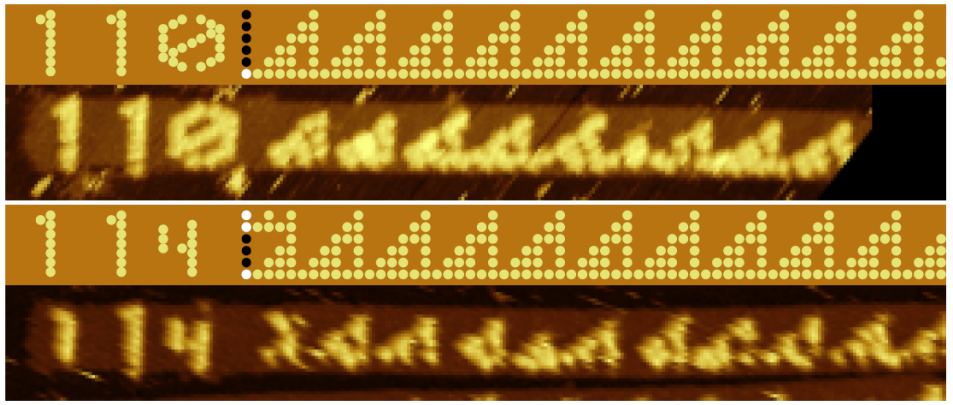
FAIRCOIN







RULE110



Counting to 63

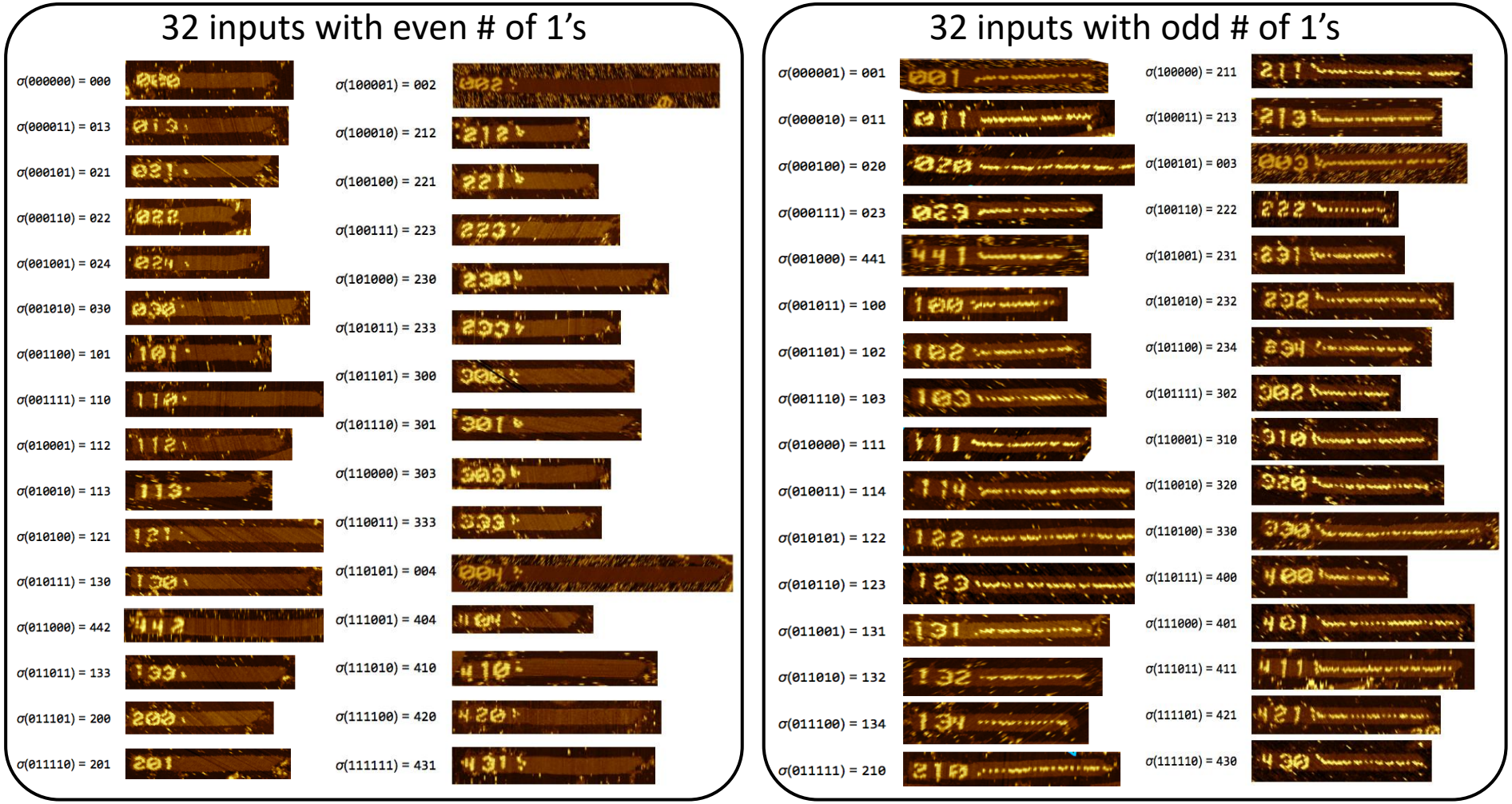
Circuit with 63 distinct strings

Is there a 64-counter?

No!
Proof by Tristan Stérin, Maynooth University
Consequence of following theorem:
No Boolean function computes an odd permutation if some output bit does not depend on all input bits.

Parity tested on all inputs

2^6 = 64 inputs with 6 bits

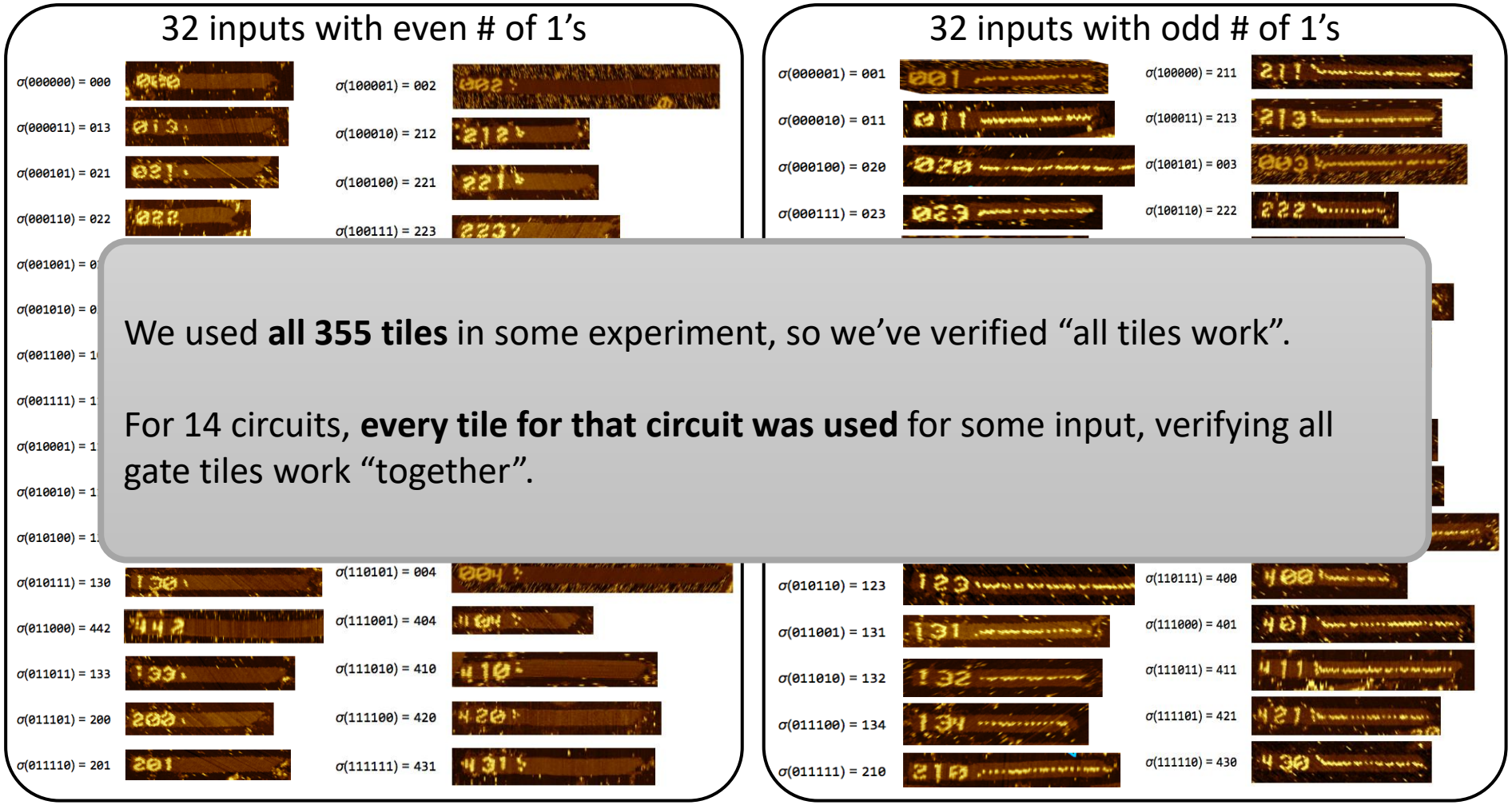


σ(6-bit input) = 3-digit barcode representing that input

150 nm

Parity tested on all inputs

2^6 = 64 inputs with 6 bits



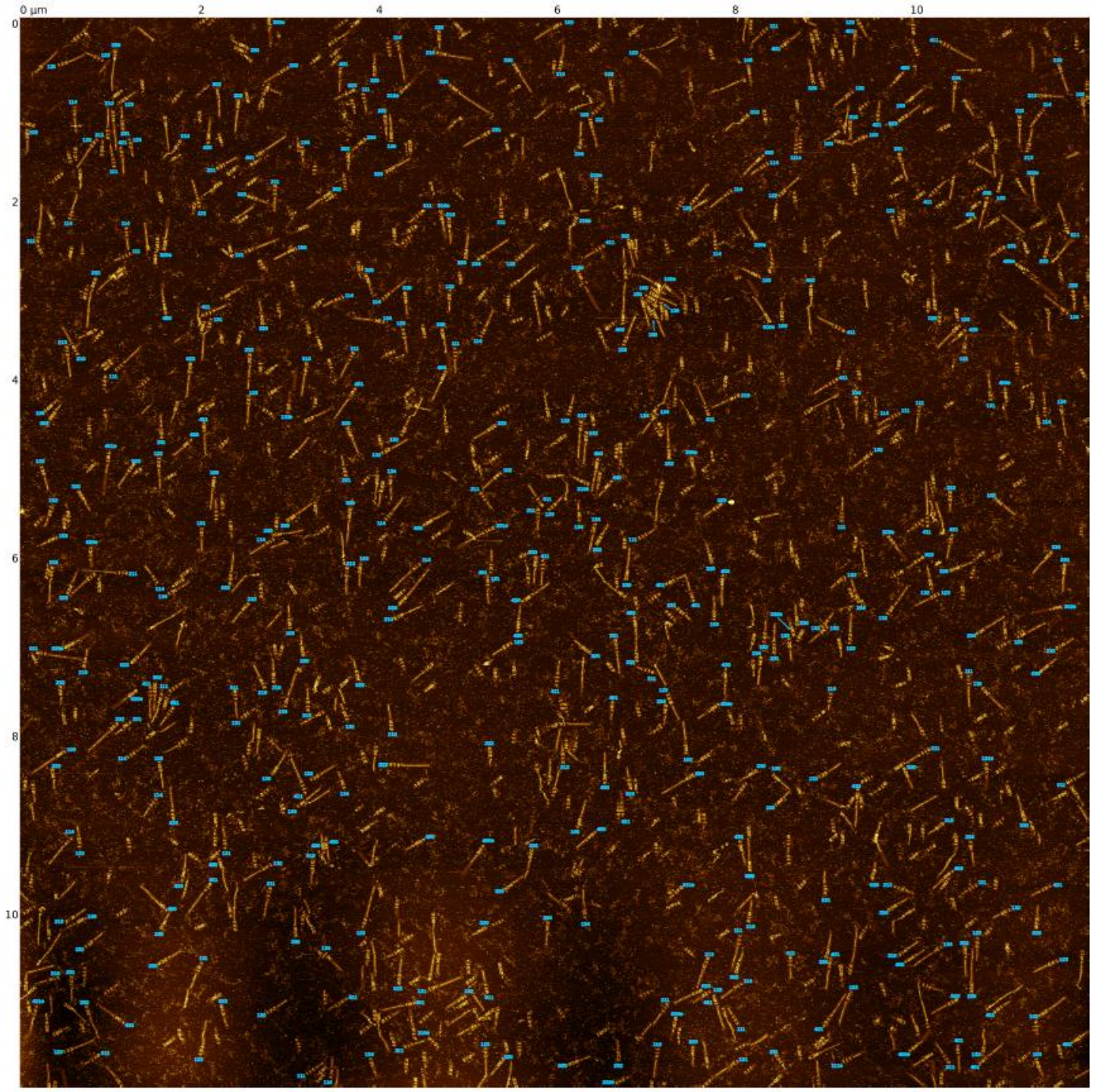
We used **all 355 tiles** in some experiment, so we've verified "all tiles work".

For 14 circuits, **every tile for that circuit was used** for some input, verifying all gate tiles work "together".

σ(6-bit input) = 3-digit barcode representing that input

150 nm

12 µm AFM image of parity ribbons for several inputs whose output is 1



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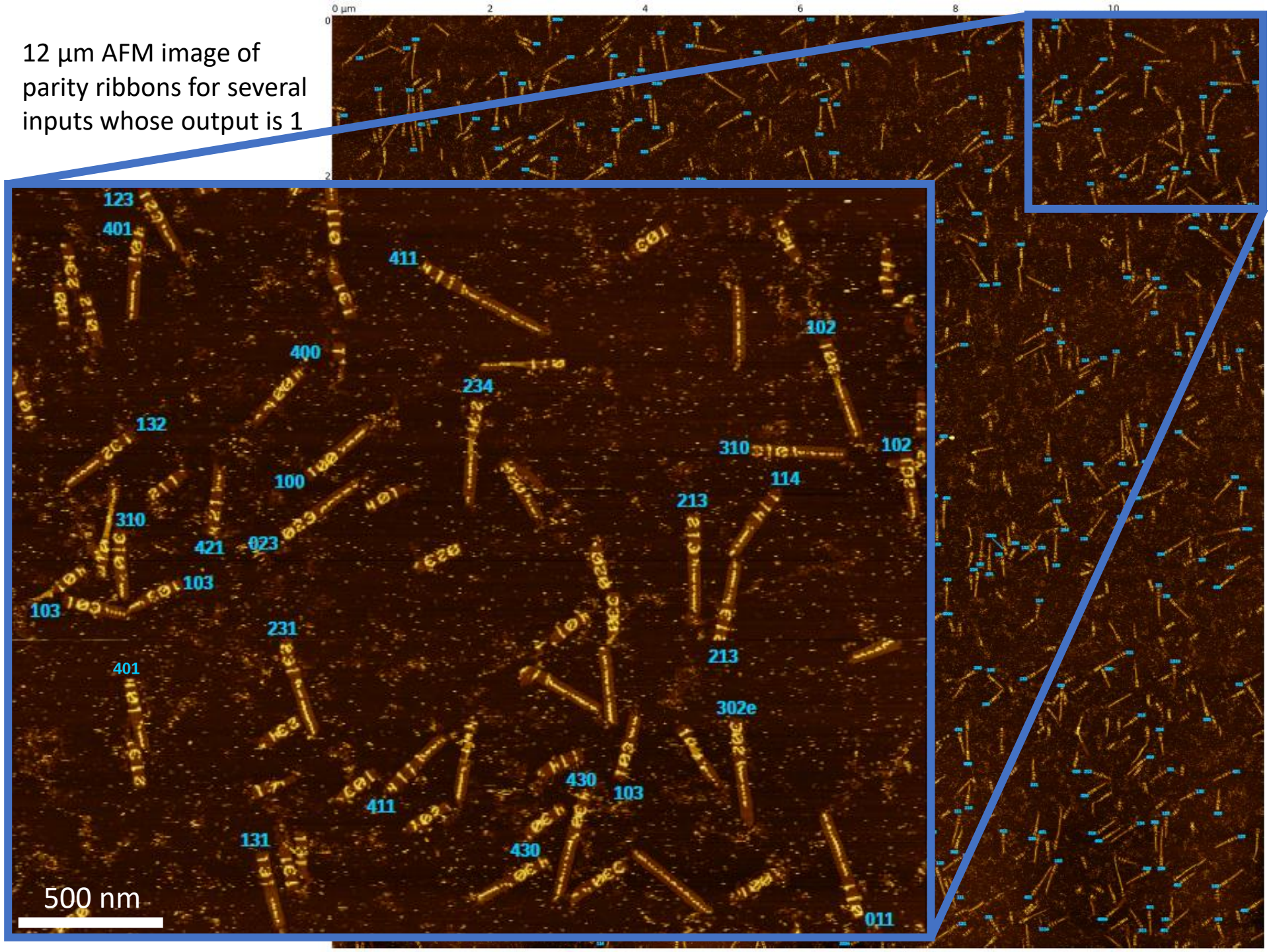

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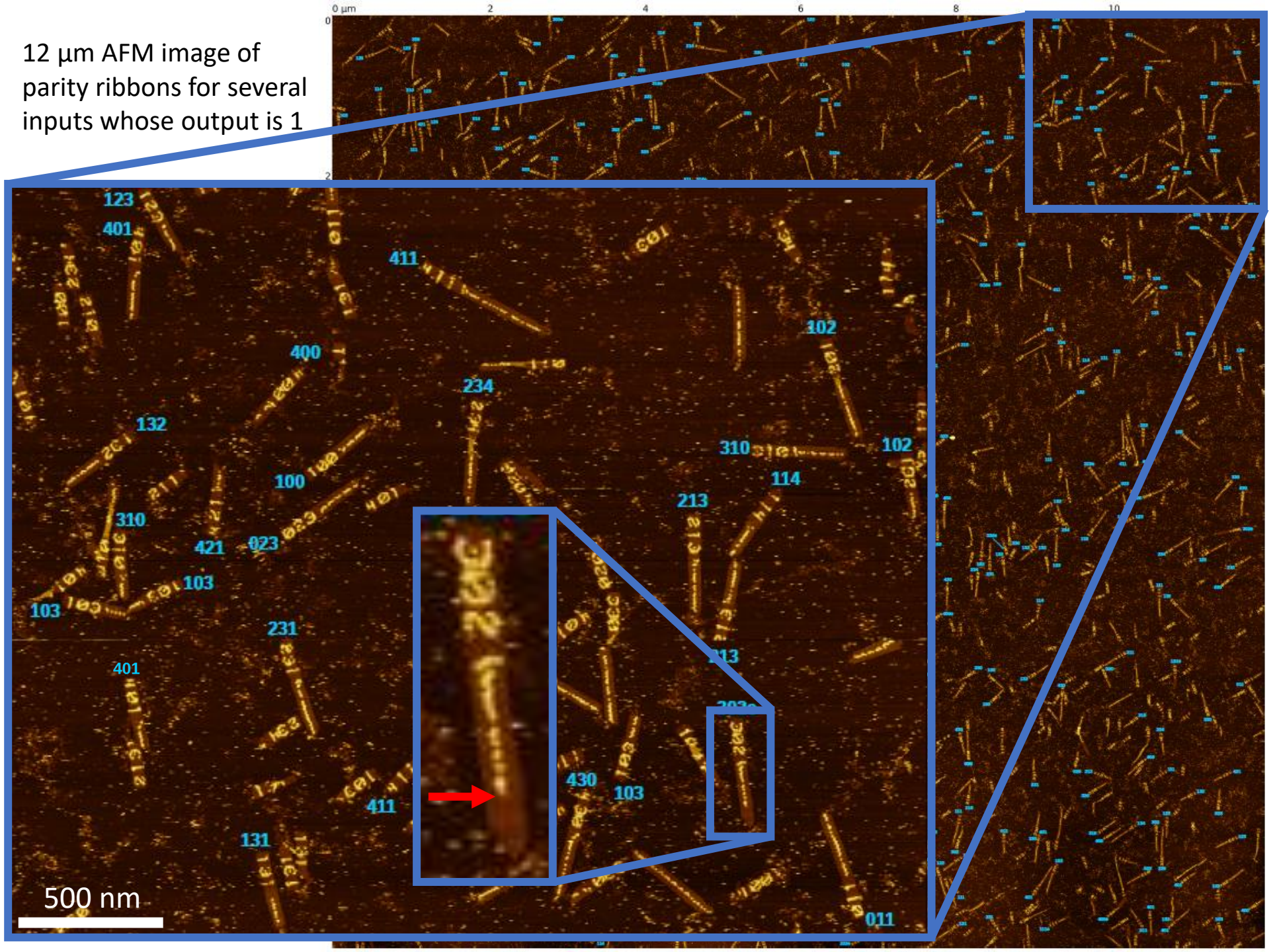

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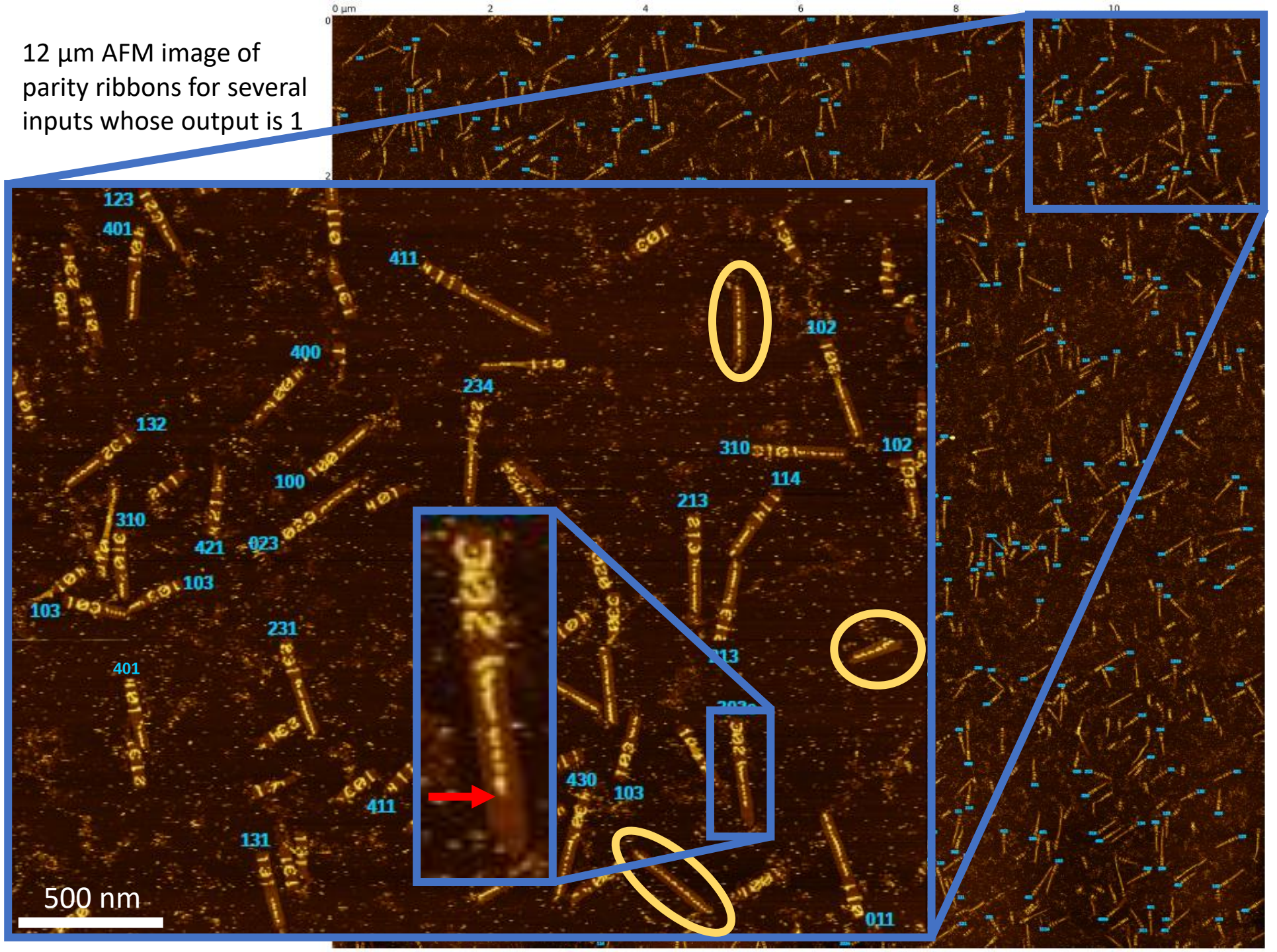

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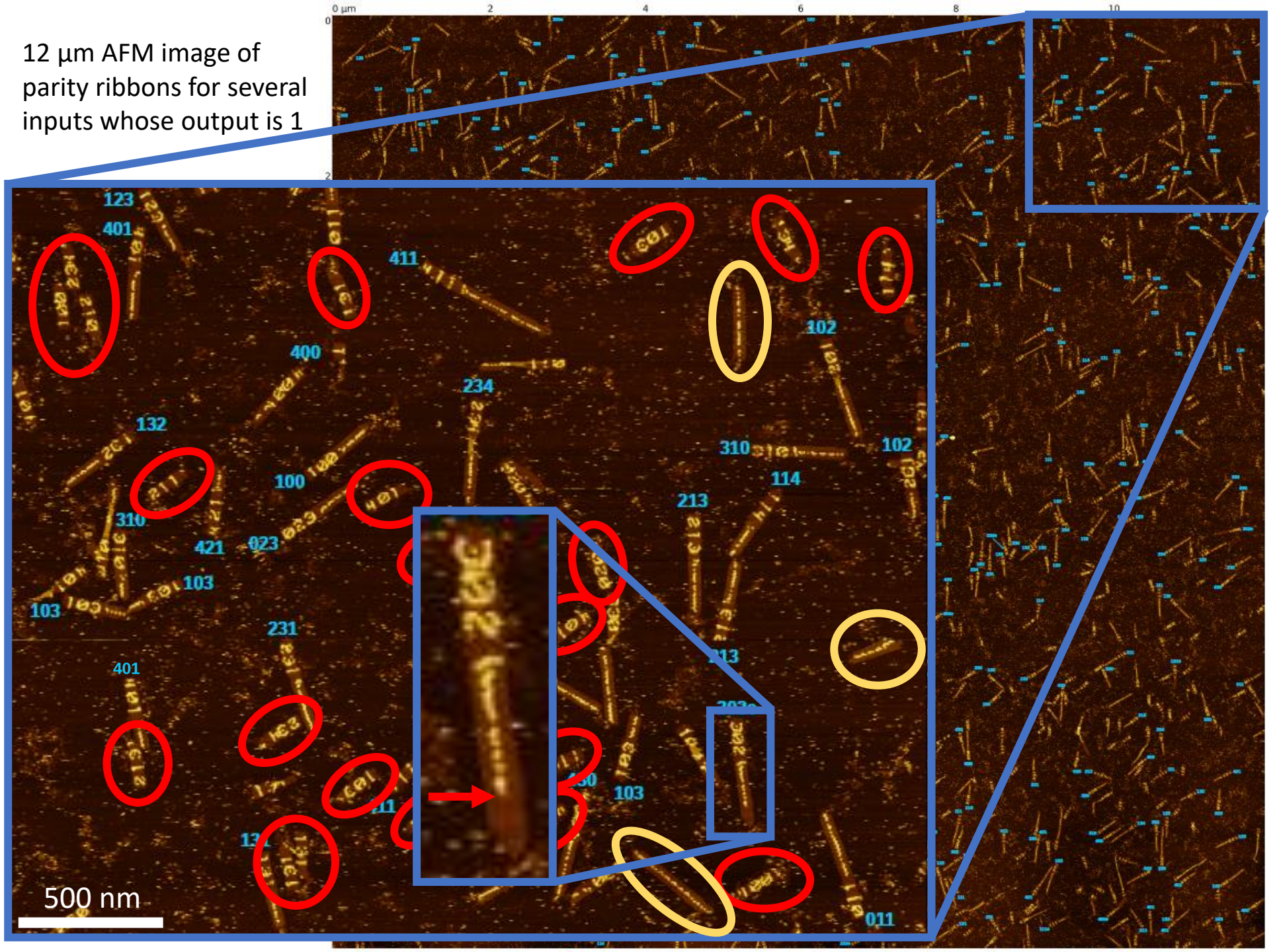

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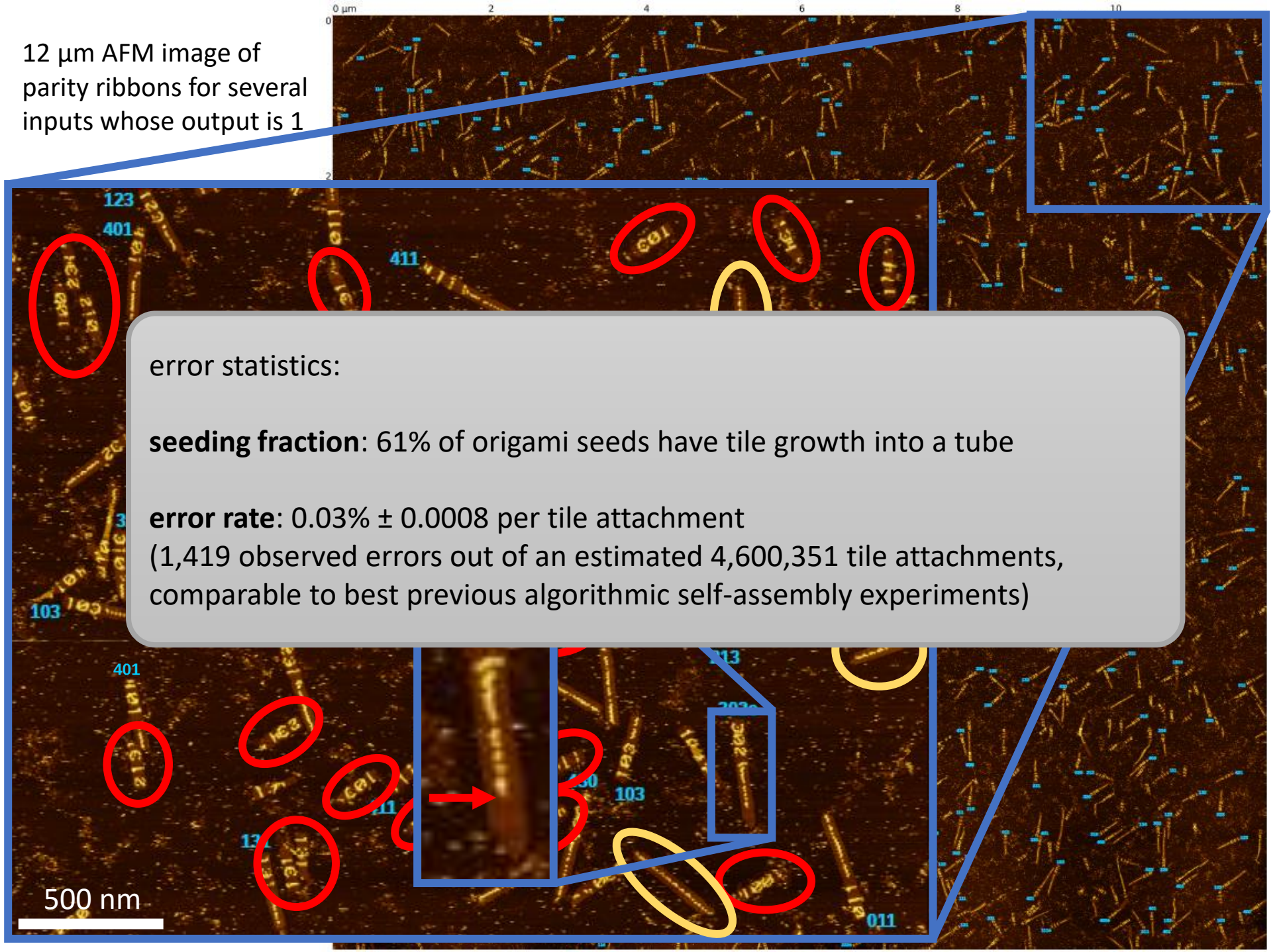


error statistics:

seeding fraction: 61% of origami seeds have tile growth into a tube

error rate: 0.03% ± 0.0008 per tile attachment
(1,419 observed errors out of an estimated 4,600,351 tile attachments, comparable to best previous algorithmic self-assembly experiments)

What did we learn?

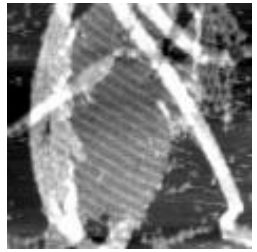
A small(ish) library of molecules can be reprogrammed to self-assemble reliably into many complex patterns, by processing information as they grow.

What did we learn?

A small(ish) library of molecules can be reprogrammed to self-assemble reliably into many complex patterns, by processing information as they grow.

Contrasting with other self-assembly work:

more algorithmic control
than **periodic** self-assembly



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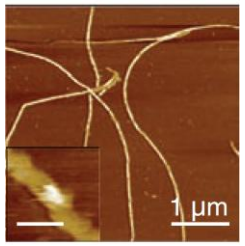

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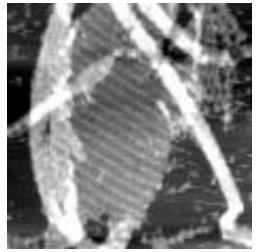

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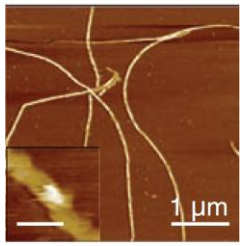

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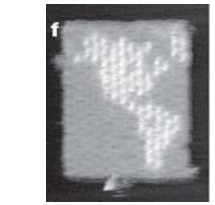

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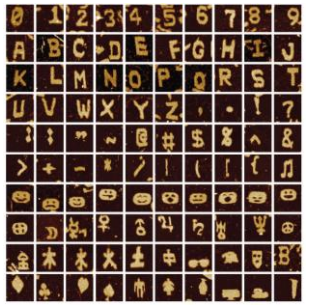

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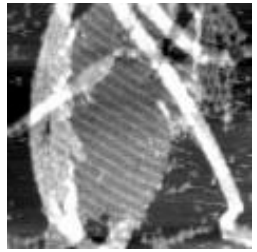

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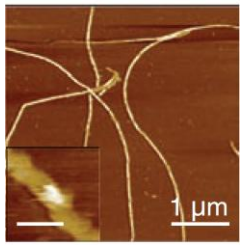

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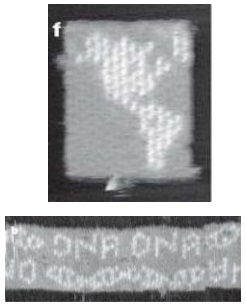

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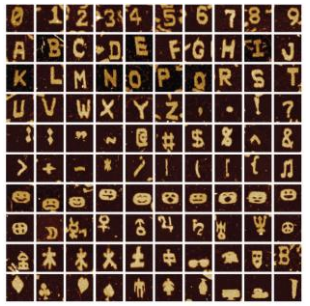

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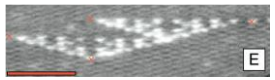
order of magnitude more tile types available than previous **algorithmic** self-assembly

double-crossover tile lattices



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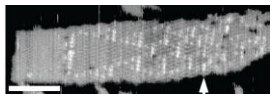

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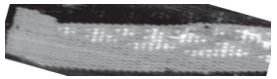

(Barish et al., *PNAS* 2009)

(Evans, *Ph.D. thesis* 2014)

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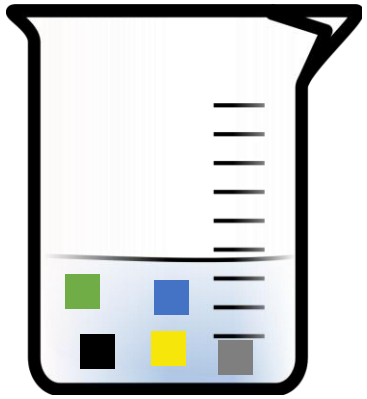

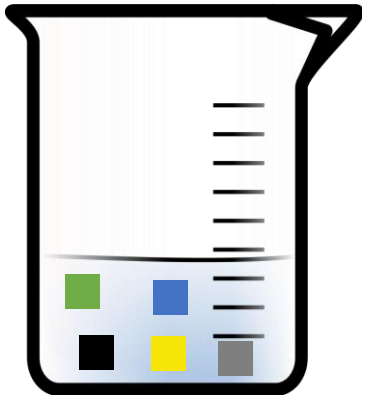

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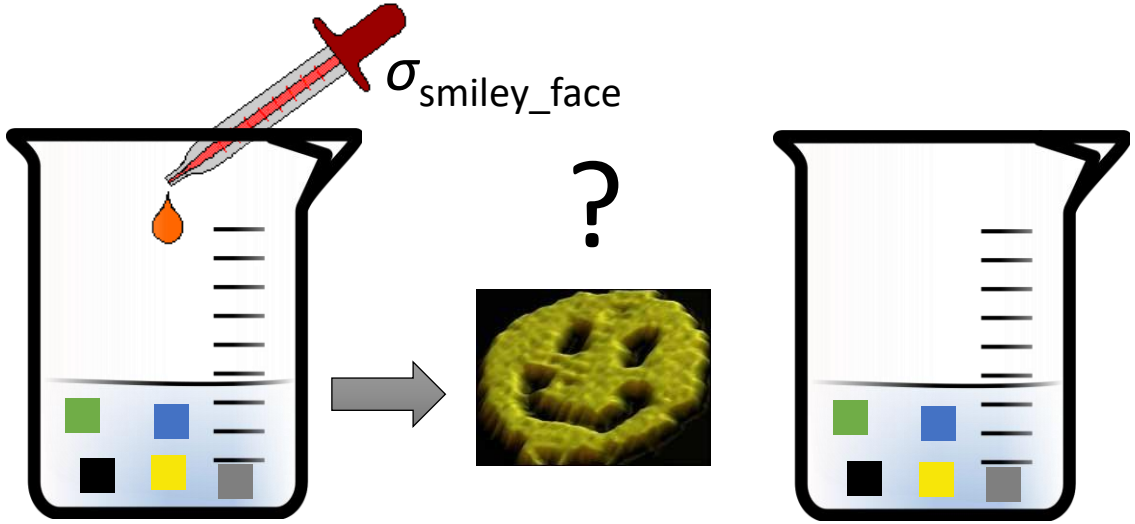

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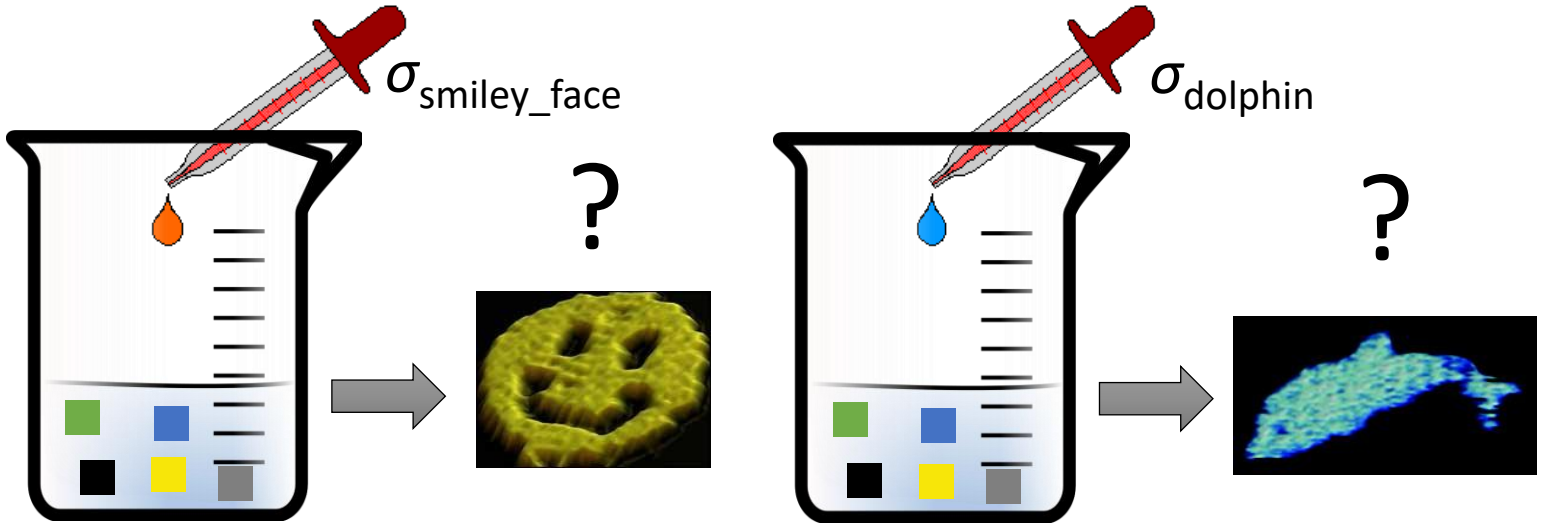

These tiles are universally programmable for building any shape.

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