Deterministic lateral displacement (DLD) is a technique for size fractionation of particles in continuous flow that has shown great potential for biological applications. Several theoretical models have been proposed, but experimental evidence has demonstrated that a rich class of intermediate migration behavior exists, which is not predicted. We present a unified theoretical framework to infer the trajectories of particles in the whole array on the basis of trajectories in a unit cell. This framework explains many of the unexpected particle trajectories reported and can be used to design arrays for even nanoscale particle fractionation. We performed experiments that verify these predictions and used our model to develop a condenser array that achieves full particle separation with a single fluidic input.

**Significance**

Deterministic lateral displacement (DLD) is a technique for size fractionation of particles in continuous flow that has shown great potential for biological and clinical applications. Several theoretical models have been proposed to explain the trajectories of different-sized particles in relation to the geometry of the pillar array, but experimental evidence has demonstrated that a rich class of intermediate migration behavior exists, which is not predicted by models. In this work, we present a unified theoretical framework to infer the trajectories of particles in the whole array on the basis of trajectories in the unit cell. This framework explains many of the unexpected particle trajectories reported in literature and can be used to design arrays for the fractionation of particles, even at the smallest scales reaching the molecular realm. We also performed experiments that verified our predictions, even at the nanoscales. Using our model as a set of design rules, we developed a condenser structure that achieves full particle separation with a single fluidic input.


**Reviewers:** M.O.M., Rockefeller University; C.S., University of Rome; and C.T., Peking University.

The authors declare no conflict of interest.

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This article contains supporting information online at www.pnas.org/lookup/suppl/doi:10.1073/pnas.1706645114/-/DCSupplemental.
A handful of theoretical models (15–17) (summarized in SI Appendix) were developed to account for the nondichotomous particle trajectories in DLD arrays (Table 1). In their explanation, previous models relied either on array geometries with restricted conditions (e.g., \( D_x = D_y \)) or on the flow structure proposed in the original theory. Questions regarding whether pillar arrays with periodicity \( N \) always have exactly \( N \) flow lanes in between gaps, or how the nature of particle trajectories (zigzag, bumping, or other types) depend on the array geometry, have never been systematically studied.

We present a theoretical framework to more universally determine the dependence of the migration angle of particles in terms of DLD array geometry parameters. Our model aims at capturing the deterministic nature of the trajectories, as applied to high Péclet number systems. We observed that the pillar array geometry distorts the fluid flow such that particle trajectories do not necessarily follow the periodicity or symmetry of the array, generating trajectories with migration angles that can have values between 0 and \( \theta_p \). Finally, we provide experimental verification of the model predictions using nanoscale DLD (nanoDLD) arrays of varying geometry. The model explains and the experimental data demonstrate that intermediate migration angles can occur in the purely deterministic regime.

### Results

**Fluid Dynamics in the Unit Cell.** We first study the velocity field in the unit cell of a 2D pillar array. Because the Reynolds number in typical DLD experiments is \(<10^{-3}\), we can model the fluid dynamics in our system using the stationary Stoke’s equations

\[
\mu \nabla^2 \mathbf{u} = \nabla p \\
\nabla \cdot \mathbf{u} = 0,
\]

where \( \mathbf{u} \) and \( p \) are the local velocity and pressure fields, and \( \mu \) is the dynamic viscosity of the solvent. We use nonslip boundary conditions on the surface of the pillars (arcs AB, CD, EF, and GH in Fig. 1A), where the component of the pressure gradient in the direction normal to the pillar surface is zero. Periodic

---

**Table 1. Models on the migration angle of DLD devices**

<table>
<thead>
<tr>
<th>Source</th>
<th>Pillar diameter(D&lt;sub&gt;p&lt;/sub&gt;)</th>
<th>( D_x/D_y )</th>
<th>( N = 1/\epsilon )</th>
<th>Modes</th>
<th>Mode transition</th>
<th>Diffusion</th>
</tr>
</thead>
<tbody>
<tr>
<td>Inglis et al. (2006) (12)</td>
<td>Point</td>
<td>1</td>
<td>Integer</td>
<td>Z, B</td>
<td>( \theta_p )</td>
<td>Binary</td>
</tr>
<tr>
<td>Heller and Bruus (2008) (16)</td>
<td>Point</td>
<td>1</td>
<td>Integer</td>
<td>Z, B</td>
<td>( \leq \theta_p )</td>
<td>Continuous</td>
</tr>
<tr>
<td>Long et al. (2008) (17)</td>
<td>Point</td>
<td>1</td>
<td>Noninteger</td>
<td>Z, MD, B</td>
<td>( \leq \theta_p )</td>
<td>Discrete, ( N ) dependence</td>
</tr>
<tr>
<td>Kulrattanarak et al. (2011) (15)</td>
<td>Physical</td>
<td>1,2,3</td>
<td>Integer</td>
<td>Z, M, B</td>
<td>( \leq \theta_p )</td>
<td>Continuous</td>
</tr>
<tr>
<td>Cerbelli (2012) (20)</td>
<td>Physical</td>
<td>1</td>
<td>Noninteger</td>
<td>Z, MD, B</td>
<td>( \leq \theta_p )</td>
<td>Discrete, ( N ) dependence</td>
</tr>
<tr>
<td>Present work</td>
<td>Physical</td>
<td>All</td>
<td>All</td>
<td>AZ, B</td>
<td>( \neq \theta_p )</td>
<td>Binary, ( D_0 ) dependence</td>
</tr>
</tbody>
</table>

\( \theta_p \), migration angle of bumping mode; \( \theta_p \), migration angle of zigzag mode; \( \epsilon \), row-shift fraction.
boundary conditions require that the velocity and pressure fields on the lines BC and FG are identical. At the inlet (HA) and outlet (DE), we used normal velocity gradient ($\mathbf{u} \cdot \nabla \mathbf{u} = 0$) and constant pressure, with the pressure $p_0$ at the inlet being larger than the pressure $p_1$ at the outlet. Here, we applied $p_0 - p_1 = 20$ Pa (Fig. 1C).

Note that we imposed no boundary conditions associated to the enclosing walls and treated our system as an unbounded infinite array.

The resulting fluid streamlines for different pillar diameters $D_0$ are shown in Fig. 1 C–F (Materials and Methods). It will be convenient to categorize the streamlines starting at the inlet line into two types: (i) "veering flow" and (ii) "direct flow," colored red and green, respectively, in Fig. 1D. We also define the "separatrix line" as a streamline dividing the veering and direct streamlines, which ends at a stagnation point on the bottom right pillar surface (Fig. 1F). Direct streamlines exit through the outlet of the same unit cell. In a unit cell, veering red streamlines below the separatrix line exit through the bottom segment BC to reenter through the top segment FG as the blue streamlines in the adjacent unit cell beneath, after which they proceed to exit at some point on the outlet line DE.

**Recurrence Map.** Every streamline represents the trajectory of a point-like, passively advected particle that enters the unit cell at some point $y_i$ on the inlet line and exits the unit cell at some other point $y_o$ on the outlet line (Fig. 1E). The distances $y_i$ and $y_o$ are measured as a ratio of $G$ along the inlet and outlet lines respectively, starting from the respective points A and D of the bottom pillars, and therefore $0 \leq y_i/G, y_o/G \leq 1$. We will denote $\eta = y/G$. This normalized coordinate defines a mapping $\eta = f(\eta)$ that uniquely assigns a point at the inlet line to a point on the outlet line, and that depends on the geometric parameters $D_0, D_x, D_y$ and $\epsilon$. Because the outlet of a unit cell is the input of the next unit cell, the map determines the position of a particle at the inlet of the next cell, given its position at the outlet of the previous cell.

This mapping is known in the theory of dynamical systems as a Poincaré first recurrence map, or recurrence map for short. Fig. 2A shows one instance of the map $f(\cdot)$ for a given geometry. The shape of the recurrence map $f$ determines the characteristics of the particle trajectory. The point of discontinuity on the $x$ axis corresponds to the position $y_i$, where the separatrix crosses the inlet line. All of the inlet points between 0 and $y_i$ correspond to veering streamlines and, because of the imposed periodic boundary conditions, map to the upper-left branch of the recurrence map. The direct streamlines (green) corresponding to inlet points between $y_i$ and $G$ map to the lower-right branch of the map. The recurrence map is not a straight line, but has curvature. In the point-like pillar array, the recurrence map is a straight line (SI Appendix).

The map can be iterated to track the trajectory of small particles over consecutive unit cells in the array for any system geometry. Fig. 2A and B shows a particle originally at position 0 being transported to position 1 in the outlet line. Identifying the outlet line of this cell with the inlet line of the next cell, we identify the equivalent point 1 on the inlet line. Iterating this procedure, the particle ultimately lands at position 9 on the inlet line, which corresponds to a veering streamline. The particle now follows the red streamline and moves to the cell below, which is represented as reentering the cell from the top (blue streamline) to end up at position 10. This process simulates a single trajectory over multiple consecutive cells as shown in Fig. 2C, where the 10-streamline segments described above as entering and exiting from the same unit cell are now unfolded as they exist in the actual array. Because the trajectories of Fig. 2C are equivalent to the trajectories of Fig. 2A, the recurrence map can therefore be used to represent any single trajectory over multiple cells as the composition of a single function.

We will call the span of a particle trajectory a “cycle,” which starts when the particle enters the unit cell after having zigzagged the previous pillar (i.e., right before entering the unit cell, the particle trajectory was a blue veering streamline in the previous cell), continues while the trajectory corresponds to direct streamlines, and completing the cycle when the particle escapes the unit cell as a red veering streamline. For example, the trace in Fig. 2C shows one cycle starting at position 0, corresponding to a trajectory that in the previous iteration had escaped the unit cell as a veering streamline (blue) and continued for 9 iterations as direct segments (green). In the next iteration, the trajectory exited the cell as a veering streamline (red) and started again as a direct segment (green).

These 10 segments constitute a cycle. We will call the number of segments in one cycle the “local periodicity” and denote it by $N^*$, as shown in Fig. 2C. This cycle can be written as follows:

$$\{\eta_1, f(\eta_1), f^2(\eta_1), \ldots, f^{N^*-1}(\eta_1)\}$$

where $f^{i+1}(\cdot) = f(f^i(\cdot))$. If the system had perfect symmetry, the local periodicity $N^*$ should be the same as the structural periodicity $N_p$ of the pillar array, as defined in the original model (12), and $\eta = f^{N_p}(\eta)$. In reality, the hydrodynamics in the unit cell are distorted such that the local periodicity may be different from $N_p$, as shown in Fig. 2A and B, where the last point (10) does not match with the $0^{\text{th}}$ position.

Because of this mismatch between the $N^*$ and $0^{\text{th}}$ positions, the local periodicity of a trajectory changes intermittently between two consecutive integers $N^*$ and $N^* + 1$. An illustration of this intermittency over multiple cycles is shown in detail in SI Appendix. We defined the pseudoperiodicity $\bar{N}$ of the average of local periodicities ($N^*$).

$$\bar{N} = \lim_{L \to \infty} \frac{1}{L} \sum_{j=1}^{L} N^*_j.$$  

Although we write the formula as $L \to \infty$, the number of cycles $L$ can be finite if the particle stops at a stagnation point. The value of $\bar{N}$ is typically a noninteger number between $N^*$ and $N^* + 1$.

We calculated the recurrence map of pillar arrays with different geometries, as well as the local periodicity over multiple cycles (SI Appendix, Fig. S2 A–D). As the pillar diameter $D_0$ increases, $f$ approaches the $y = x$ line, and as a result, the local periodicity of each cycle increases, even though the structural periodicity $N_p$ is the same. As $D_0 \to D_p$, the map approaches a tangent bifurcation, which is one of the known routes to chaos through intermittency.

**Geometric Effects on the Migration Angle.** The pseudoperiodicity is the average of the local periodicity of a particle trajectory over multiple cycles. Therefore, the migration angle of a trajectory can be calculated from its pseudoperiodicity. If the pseudoperiodicity is larger than $N_p$, the average trajectory direction does not fall in line with the $x$ axis of the array (Fig. 3, red and blue line). In this case, we define the trajectory as in an “altered zigzag mode,” and a nonzero migration angle is given by

$$\theta = \tan^{-1}\left(\frac{(\bar{N} - N_p)D_x}{N N_p D_y}\right).$$

The structural periodicity of the array $\theta_p$ can be computed as

$$\theta_p = \tan^{-1}\left(\frac{D_y}{N_p D_x}\right).$$

Therefore, the ratio between the altered zigzag mode migration angle and the pillar structural angle, assuming small angles, becomes:

$$w = \frac{\theta}{\theta_p} \sim \frac{\bar{N} - N_p}{\bar{N}}.$$
As $\bar{N} \geq N_p$, the altered zigzag migration angle can be greater than zero.

We computed the particle trajectories over multiple pillar arrays with the same structural periodicity, but different pillar diameters (SI Appendix, Fig. S2). As the pillar diameter increases, the pseudoperiodicity increases regardless of the initial position (SI Appendix, Fig. S3 A–D) and a corresponding increase in the migration angle results. This nonzero altered zigzag migration angle is solely the result of the geometry of the pillar array modifying the fluid flow, and not the interactions of the particle with the pillars.

In Fig. 4 A, we plot the normalized migration angle $w$ as a function of the ratio between the pillar diameter $D_0$ and the lateral pitch distance $D_y$. We can see that there is no dependence of $w$ with $D_0/D_y$ for arrays with $D_x/D_y = 2$, and $w \sim 0$ as in the original theory. For $D_x/D_y = 1.5$, we can see a mild dependence of $w$ with the normalized pillar diameter. However, at $D_x/D_y = 1$, there is a dramatic increase of $w$ with $D_0/D_y$, where the larger pillar diameters and closer pillar row spacing enhance the normalized migration angle closer to bumping mode.

To test these predictions, we experimentally measured the migration angle of 50 nm fluorescent polystyrene beads in a series of nanoDLD arrays of varying $D_0/D_y$ (Materials and Methods). These experiments used full-width injection, in which particles are introduced across the entire width of the array inlet, and the migration angle is measured based on how much the particle flux is deflected (11). Beads were run at velocities $>5$ mm/s (toward the maximum obtainable with our experimental setup) to reduce the influence of diffusion ($Pe \sim 200$).

Importantly, for all gap sizes used, the original theory (12) would predict a critical diameter around 80 nm, and, therefore, according to this theory, 50-nm particles should follow a zigzag mode. Fig. 4 B–D shows fluorescent microscope images of deflected bead flux for different $D_0/D_y$, showing how the geometry of the array is influencing the lateral displacement of the beads. Fig. 4 A plots the experimental results along with our model predictions.
Although there is considerable agreement between theory and experiment, the agreement is not perfect. Potential reasons for this discrepancy include the fact that the pillar array in any real device is not infinitely periodic, so that boundary effects could influence \( \theta \), as well as effects of the 3D nature of the flow, and the inability to completely nullify particle diffusion with high-velocity flow.

To verify the predicted dependence of the migration angle with \( D_x/D_y \), we fabricated and tested a nanoDLD device with two sequential pillar arrays with ratios \( D_x/D_y \) of 1 and 2, and \( D_0 = G = 200 \) nm, and \( D_y = 400 \) nm (Fig. 4E). Fig. 4F shows a fluorescence microscope image mosaic of 50-nm beads injected into the full width of the device with a first array of \( D_x/D_y = 1 \) and 250-\( \mu \)m length, and a second array of \( D_x/D_y = 2 \) and 500-\( \mu \)m length. Beads entering the first array follow the altered zigzag mode and deflect laterally to the upper wall (migration angle \( \theta = 2.86 \), normalized migration angle \( w = 0.5 \)). When the deflected flux reaches the second array, the increased axial pitch reduces the altered zigzag mode, and the bead flux no longer deflects significantly (migration angle, \( \theta = 0.36 \); normalized migration angle, \( w = 0.12 \)). This phenomena is in contrast with the original theory (12), according to which the critical particle diameter should be 80 nm, and therefore 50 nm beads should be in zigzag mode for both regions of the array.

Our model predicts an important phenomenon—the deflection of small particles with high \( D_0/D_y \) ratio that should operate in zigzag mode according to the original theory, a feature with practical utility. The structure in Fig. 4G exploits this concept to achieve high-efficiency separation from a single fluidic input. In the first part of the device, which we refer to as a condenser, \( D_0/D_y \) ratio should be higher than 0.7 and both 100nm and 50nm particles are deflected without the requirement of smaller gaps to bump the 50 nm particles.

Fig. 4. Experimental validation of our predictions using nanoDLD arrays. (A) Normalized migration angle as a function of the pillar-to-pitch ratio for both simulations and experiments. B–D and F–G points represent the corresponding experiments in A–C, F, and G. (B–D) Fluorescent microscope images of the array outlet of nanoDLD devices with increasing \( D_0/D_y \) and \( D_x/D_y = 1 \). The bright regions are fluorescent bead (\( D_p = 50 \) nm) fluxes, which have been deflected by different degrees to the right-hand wall. The experimental details are presented in Materials and Methods. (E) The microscope image of the nanoDLD array with a heterogeneous structure (\( D_x/D_y = 1 \) and 2). (F) The fluorescent image of a 50-nm particle migrating through two regions of a channel. The deflected particle trajectory lays between the bumping and zigzag modes (green and red lines). F. Inset provides a SEM image showing heterogeneous nanoDLD structure at the boundary of two regions. (G) A heterogeneous structure for highly efficient separation. The diagram shows the full-width injection trajectory of the 50 nm and 100 nm particle. The fluorescent images are composites of separate particle measurements aligned with the representative diagram.
particles. In the second part or sorter region, $D_p/D_y = 0.4$ and the array fractionates the particle mixture in the opposite direction. Together, the condenser and sorter arrays in series greatly simplify the fluidic input, while still allowing complete separation of particle populations by using a full-width injection layout.

**Pillar-Size Effect on the Critical Diameter.** We can use the recurrence map to model the trajectories of finite-size particles and study the dependence of the critical diameter on pillar geometry. As in Inglis et al. (12), our model uses two simplifying assumptions: (i) Flow is not modified due to the particle’s presence; and (ii) the hard walls and our finite size particles interact with perfect inelastic collision.

We simulated the trajectories of the particles’ center of mass with different diameters $D_p$ within a unit cell. As the particle size increases, only the trajectories starting between $D_p/2$ and $G - D_p/2$ are allowed at the inlet line due to the hard-wall repulsion. When the particle is close to the pillar surface, several trajectories starting at different positions collapse to one trajectory [a phenomenon known as directional mode locking (21)], as shown in Fig. 5 A and B. When the particle diameter is less than the critical diameter $D_c$, there exist veering streamlines (red lines in Fig. 5 A), but with larger particles, only direct streamlines remain (green line in Fig. 5 B). In this case, all trajectories collapse, leaving only direct streamlines, meaning that only a bump mode remains.

Fig. 5 C and D show the recurrence maps for particles with different diameters. With increasing particle size, the allowed region in the map (bright gray) shrinks due to geometric constraints of the particle and pillars. When the starting position is close to the streamline that leads to the stagnation point ($y_s$), the mapping function becomes constant because the collapsed trajectories give the same final position. When a particle has a larger radius than the distance to the streamline that leads to the stagnation point ($D_p/2 > y_s$), the map will have a fixed point, and all trajectories will collapse into the bumping-mode streamline (Movie S1). Therefore, the recurrence map gives the critical diameter of the system.

$$D_c = 2y_s$$

Note that this $y_s$ corresponds to the width of the $1/N$ flux stream in the original theory (12).

The critical diameter dependence on the geometry is shown in Fig. 5 E for various $D_x/D_y$ systems. Unlike the original theory (dashed line), the critical diameter in our model depends on the ratio $D_0/D_y$. We observe two different behaviors depending on the ratio $D_x/D_y$. When the $D_x/D_y > 1$, the ratio $D_c/G$ decreases as the pillar size increases (red line), meaning that $D_c$ will be smaller than suggested by the original theory. When $D_x/D_y = 1.5$ and 2 (green and blue lines), the critical diameter increases as the pillar size increases. These results can be understood based on the particle trajectory’s pseudoperiodicity. The inlet flow is divided by $N$ lanes separated by streamlines that will end in a stagnation point downstream. The critical diameter can be calculated as twice the width of first lane, which carries $1/N$ of the total flux through the inlet. In the original DLD model,

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**Fig. 5.** The trajectory of particles with different diameters and corresponding recurrence maps. (A and B) Particle trajectories inside a unit cell with different particle diameters. Here, the particle diameters are $D_p = 20, 45$ nm, respectively, and $N_p = 10, D_0 = 240$ nm and $D_x = D_y = 400$ nm. A black circle represents the particle with a given radius. (C and D) Corresponding recurrence maps for a particle starting at the midpoint ($y_s = 0.5$). The dark gray regions represent a forbidden region by the particle–wall interaction as directional locking. The black circle shows the particle radius with a scale of gap size. (E) The critical diameter dependence on the pillar geometry ($D_0/D_y$). (F) The effect of DLD array geometry on the transition from altered zigzag to bumping mode. The plot shows the normalized migration angle as a function of particle size for different pillar diameter ($D_0/D_y$). The simulated particle diameters $D_p$ range from 1 to 110 nm, and the pillar diameters $D_0$ are 80, 120, 160, 200, 240, 280 nm. The dotted lines represent the critical diameter calculated from $E$. 

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the periodicity $N$ is the structural periodicity $N_p$ of the array, whereas in our model, $N$ increases substantially with the pillar diameter when $D_p/D_p = 1$. Therefore, the first stream width decreases with large pseudoperiodicity $N$, and, as a result, the critical diameter decreases. A similar argument can be used to explain the critical diameter dependence on the pillar size in the case of $D_p/D_p = 1.5$ and 2, whereas $N \sim N_p$.

Fig. SF shows the transition between the altered zigzag mode and bumping mode for diameters $D_p$, ranging from 1 nm to 110 nm, and for different pillar diameter $D_p$ and gap size $G$. The migration angle shows a binary transition between a zigzag and bump mode in all cases, as expected when diffusion is not present. Consistent with our earlier observations, the migration angle of small particles increases, as the pillar diameter increases, and the critical diameter decreases as the pillar diameter increases. Interestingly, the migration angle for particle diameters $D_p < D_p$ has subtle transitions for $D_p = 80, 120, 160$ nm. These transitions may be related to the intermittency of the local periodicity similar to a devil's-staircase-like structure in a phase-locking system. (22)

Conclusions

The original theory explaining the principles of operation of DLD arrays was formulated under the assumption that the symmetry of the fluid streamlines would match the symmetry of the pillar array. Our work shows that this assumption does not provide a complete picture of the underlying physics. By reformulating the principle of operation of DLD arrays, we have shown that particle trajectory dependencies on array geometry are stronger than in the original theory and that the particle trajectory behavior is considerably richer than originally thought. The nonzero migration angle of small particles discussed in this work stems from the anisotropic permeability of row-shifted parallelgram DLD array layouts, as has been discussed by other authors (15, 23). Indeed, we confirmed that the velocity field averaged over a unit cell has a nonzero lateral component and that the angle between the average velocity vector and the pressure gradient direction is almost identical to the migration angle of small particle trajectories calculated by using the recurrence map. (See Appendix, Fig. S1).

Despite the remarkable agreement between theory and experiment shown in Fig. 4, we should note the assumptions and limitations of our theory. First, we assume an infinite lattice and neglect the effect of the array-bounding walls. Second, the fact that the mean direction of the streamlines follows an angle not in the direction of the pressure gradient imposes an anisotropic permeability on the fluid flow. In finite arrays, this off-axis flow would create a depletion of fluid from one wall and a buildup of fluid on the other wall, modifying the pressure boundary conditions assumed in our model. Both of these limitations will be less severe if the array is sufficiently extended. A third limitation is the assumption that the unit cell has constant pressure at the input and output. Simulations done in systems of two or more unit cells show that this assumption is approximately correct. Finally, we assume that the particle presence does not affect the streamlines and that the particle–pillar interaction can be reduced to a hard-wall repulsion. These assumptions can be justified by using Maxey–Riley equations and the fact that the Reynolds number in the system is small (supplement in ref. 11). (Note that some of these approximations are also made in the original theory.)

Our theory provides rules on how to modify the pillar array geometry to tune not only the critical diameter, but also the migration angle of small particles in the altered zigzag mode. These results can be used as guiding principles for DLD design to control the separation of particles and biocolloids from the microscale to the nanoscale. The condenser adaptation of these design rules also provides a path toward simplified DLD systems that can achieve full separation of particles by using a single fluidic inlet.

Materials and Methods

Continuum Simulation. We calculated the 2D velocity and pressure fields in a unit cell by solving the Navier-Stokes equation using the finite volume method. Several techniques, such as the finite element method (COMSOL) and the Boltzmann lattice method have been used to simulate particle motion in a pillar array (15, 24). In this work, we used OpenFOAM together with its Lagrangian particle tracking library (25). In addition, gmsh was used for a mesh generation tool. By using it, the geometric components ($D_p, D_p, \rho$) were parameterized, and different designs were simulated in a single code (26). The convective terms were approximated using an unstructured mesh with a total of $\sim 50,000$ mesh cells. We explored different mesh sizes as well as different residual resolutions, confirming the consistency of the simulation results. The residual resolution for the simulations shown in the work was set as $10^{-6}$.

A discrete element method was used to calculate the particle trajectories in the fluid, taking into account the necessary particle–pillar interaction. For the latter, we used a simple hard-wall repulsion model with homemade code modifying OpenFOAM's Lagrangian particle library. Other particle–pillar interactions such as soft-wall models are available, but the simplified model can grab the essence of the dynamics (21). We calculated and mapped the trajectories of a 1 nm diameter particle as a streamline tracer at 1,000 starting positions along the segment AC in Fig. 1. These particle traces are almost identical to the flow streamlines. An average fluid velocity of 300 $\mu$m/s was used, and different time steps were tested to confirm the consistency of the simulated trajectories. The recurrence map was generated by using Python as the programming language. The continuous line in the map $f(x)$ was constructed by linear interpolation between 1,000 ($n, f(x)$) data points. We also confirmed that the results of the recurrence map with >1,000 points were consistent with the simulations using 1,000 points at the inlet.

Chip Fabrication. The nanoDLD device arrays were fabricated on 40-× 40-mm chips on 200 mm wafers to permit fluorescence imaging of nanoscale polymer beads as described in ref. 11. Briefly, lithographic definition of fluidic features were first etched into an 150 nm-thick SiO$_2$ hard mask (HM) by using a two-stage process: (i) optical contact lithography was used to define the microchannel features followed by a reactive-ion etch (RIE) to transfer these features into the SiO$_2$ HM; and (ii) after stripping the resist from (i), a combination of electron-beam (e-beam) and deep-ultraviolet (DUV) lithography were used sequentially to pattern the nanoDLD pillar array features by adjoining the microchannels, transferring these features into the Si substrate using a single RIE process. With both features etched into the HM down to bulk Si, all open features were simultaneously etched into $\sim 400$ nm into the silicon by using an DPSII ICP etch chamber (Applied Materials). This double-stage lithographic process permitted predefinition of microchannels, and subsequent use of e-beam/DUV lithography provided a robust platform for rapid and flexible prototyping of various nanoDLD devices within this single chip fabrication process. The HM oxide in 10:1 dilute hydrofluoric acid and regrowing a thin thermal oxide layer ranging from 10 to 50 nm, depending on the desired gap size.

The 40-× 40-mm chips were bonded with 170-μm-thick borosilicate coverslips, micromachined with a custom array of holes (Mark Optics Inc.) for making fluidic connection between the chip and an external connector. Chips were sequentially cleaned with acetone and then isopropanol and $N_2$ jet-dried. To prepare a hydrophilic bonding surface, chips were run in O$_2$ plasma RIE for 20 min, and then both the chips and coverslip were run in Piranha solution [1:1 (vol:vol) sulfuric acid:nitric acid, 90 °C for 60 min]. Chips and coverslips were then washed sequentially with deionized water, acetone, and then isopropanol and immediately $N_2$ jet-dried. The coverslip was then immediately aligned and contacted to the chip surface. Pressure, applied by a tweezer tip, was used to make a tacking bond between the coverslip and chip, in which pockets of air were pushed out of the interface. Bonded chips were then annealed at 550 °C for 8 h.

Priming Chip. To wet the chips a 100-mL solution of 2% (vol:vol) Tween 20 (Sigma-Aldrich) in deionized water was prepared in a 500-mL beaker. A custom glass holder was used to position the chips vertically, with the inlet ports down, in the Tween 20 solution to allow capillary wetting of the fluidic inlet. Typical inlet pressures were $\sim$ 1 bar; the liquid to reach the outlet ports. The chip was then fully submerged in Tween 20 solution for 16–24 h to allow any residual bubbles to clear from the device. Chips were then rinsed thoroughly with deionized water and $N_2$ jet-dried. Immediately, immediately
the chip was transferred to a custom-built flow cell, and the inlet ports were filled with Tween 20 solution to prevent drying out of the fluid channels. For small gap sizes (e.g., G = 120 nm), some bubbles would remain in the fluid channel near the array and would have to be purged before running experiments. For purging method, the flow cell was connected to the drive pump (vida infra) and run at 10 ~ 14 bar for 10 ~ 30 min, during which time all remaining bubbles collapsed.

**Bead Displacement Experiments.** NanoDLD device chips with gap sizes ranging from G = 120 ~ 250 nm were prepared as described above. Fluorescence-dyed polystyrene beads with carboxylate groups (Bangs Laboratories Inc.) were used as the testing sample, having particle diameter $D_p = 50 \text{ nm}$, 1.0% (g/mL, 1.4 × $10^{-14}$ particles per mL, with excitation at 488 nm and emission at 510 ~ 520 nm. The 1:100 (vol/vol) dilutions of as-supplied bead stock to 2% (vol/vol) Tween 20 solution were prepared and used for all experiments. Diluted bead solution was loaded into the fluid cell (10 μm), injected at 10 bars, and watched via a microscope/camera until the beads arrived. For driving the fluid, a syringe pump and pressure sensor (QMmix, Cetoni GmBH) was connected to the inlet port of the fluid cell. Imaging was carried out on a Zeiss Scope.A1 upright fluorescence microscope coupled with an Andor iXon Ultra 897 (Andor Technology Ltd., Oxford Instruments) EMCCD camera connected to a computer, where both imaging and pump pressure were controlled. Exposure time was 17.9 ms per frame and 200 frames per video. A 470 nm LED was used for excitation, with the Zeiss filter set 38 HE (470/40 excitation, 495 beam splitters, 525/50 emission). Once bead flow across the entire array width was established, the pressure was increased to 18 bars (the maximum permitted by our system), and the bead fluorescence was imaged along the length of the array. The bead velocity was estimated from extrapolation of the speed measured from particle traces as a function of applied pressure, 0.1 ~ 1 bar. The fluorescence profile at the outlet of the array was measured by using custom software, using the average intensity of video of the bead flow. From the fluorescence intensity profile, the displacement of the bead flow, $\Delta W$, was estimated from the inflection point at the onset transition from no fluorescence (depleted region) to fluorescence (bead flux region). The bead migration angle was then calculated from the displacement by $\theta = \tan^{-1}(\Delta W/L)$, with the length of array, L = 510 μm.

**ACKNOWLEDGMENTS.** We thank Stacey Gifford, Navneet Dorgra, and Pablo Meyer from the IBM Nanobiotechnology program, and Prof. Howard A. Stone from Princeton University for fruitful discussions. We also thank the IBM Microelectronics Research Laboratory staff for processing the microfluidic chips used in this study, with special thanks to Simon Dawes and Markus Brink for e-beam lithography assistance.

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Movie S1. The concept of the recurrence map can be more clearly understood by using an animated description of the dynamics. Our animation shows the time evolution of a particle trajectory and connects it to the corresponding dynamics in a recurrence map. In the movie, the three panels from the left show the trajectory in multiple pillars, the trajectory in a unit cell, and the recurrence map, respectively. Here, $N_p = 5$ and the pillar diameter $D_0$ is 200 nm. The pillar-to-pillar distance ($D_x = D_y$) is 400 nm, so that the ratio ($D_0 / D_y$) is 0.5. The long trajectory is composed of eight total transition segments starting from 90% of the pillar gap ($\eta_0 = 0.9$). For the particle with radius larger than the critical diameter ($\eta_c - \eta$, the trajectory is similar to that of the small particle until it veers around the pillar for the first time. However, because of the shift of the center of particle by the pillar repulsion, it cannot veer around the pillar and bumps on the post surface. After it follows the closest possible streamline to the separatrix line, it ends up to the same initial position [the directional locking (21)]. The animation in the middle shows this transition by overlaid trajectories inside a unit cell. In the recurrence map, the mapping $f$ intersects the identity function ($y = x$), which becomes a fixed point in the dynamics. Here, the blue circle indicates the initial position at the inlet, and the red circle indicates the final position at the outlet.

Movie S1

Other Supporting Information Files

Si Appendix (PDF)
Supplementary Information Appendix

Previous models

A handful of theoretical models were developed to account for the non-dichotomous particle trajectories in DLD arrays (Table 1 in main text. Note that the table only considers stretched array designs and deterministic dynamics. Tilted square arrays are not within the scope of this paper). Diffusion could be a source for displacement angles between $0^\circ$ and $\theta_p$. Heller and Bruus proposed a model that included diffusion and particle size dispersion, which reproduced the continuous change in migration angle as a function of particle diameter. They assumed that particles diffuse as they are advected by the flow in an array of point-like pillars. Heller and Bruus’ theory, however, is not directly relevant to the deterministic dynamics studied in this paper.

Interestingly, non-diffusive dynamics can also give rise to particle trajectories that are neither zigzag nor bumping. Long et al. discovered another non-binary deterministic migration angle in the case where the row-shift ($\epsilon$) is such that $MD_y = N\epsilon$ with $M$ and $N$ co-primes. Long et al. called this non-binary behavior multi-directional sorting modes, and showed the existence of 3 or 4 different migration angles depending on $M$ and $N$. In these models the pillars were treated as points, ignoring the flow distortion due to the finite pillar diameter (Figure 1C-F). Cerbelli also studied rational row-shifts in the case of pillars with finite size and also found multi-directional modes.

Kulrattanarak et al considered finite size pillar arrays with different geometric parameters $D_x$, $D_y$ and $N$, and observed (numerically and experimentally) particle trajectories which they named “mixed motion”, with angles in between zigzag and bumping modes. According to the authors, mixed motion (which Kulrattanarak et al observed when $D_x/D_y \leq 2$) was associated with a symmetry breaking in the width of the two flow lanes at the two opposing pillars that define a gap. While this work provided a significant step forward in capturing the phenomenology of DLD with realistic geometries, it did not provide an explanation of how the symmetry breaking of the fluid lanes leads to mixed motion in specific geometries. Importantly, these authors considered the streamlines and separatrices as having the same symmetries as the pillar array, as proposed in the original theory.

Anisotropic permeability of a stretched DLD array

The problem studied in this paper is a pressure-gradient driven fluidic system in a perfectly periodic lattice of pillars. For these kinds of systems, several studies have shown that the micro-structure of the lattice can cause anisotropic permeability, where the flow direction is not aligned with pressure gradient direction. Therefore, small particles advected by the flow can follow trajectories with non-zero migration angle. When wall-bounded arrays are considered instead of an idealized infinite lattice,
the external walls impose additional boundary conditions. Some authors have assumed that the presence of the walls force the flow to have no lateral velocity (that is, zero average velocity perpendicular to the walls) in each unit cell, an assumption that results in the impossibility of anisotropic permeability. However, recent experiments (included our own results in this paper) have shown that the dynamics of passively advected particles in wall-bounded stretched DLD arrays can exhibit a lateral velocity and therefore anisotropic permeability.

In the nanoDLD array considered in this paper, there are 125 pillars in between the walls delimiting a 50µm wide channel, and 1250 pillars in the axial direction between the inlet and outlet of the array. Whether the assumption that channel-bounding walls (which impose a global boundary condition) force a zero lateral velocity in each internal unit cell (a local flow pattern) is valid, or if the number of pillars within walls is large enough to be well approximated as an infinite array, can be addressed experimentally. Figure 4A of the main text show that small particles indeed have non-zero migration angle, consistent with the predictions of our model that assumes an infinite array. This agreement is then expected when the ratio of the width of the array (W) to the lateral pitch (D_y) is sufficiently large, that is, when W/D_y >> 1.

In the main text we studied the migration angle of particles in DLD arrays in terms of the local periodicity and pseudo-periodicity of individual particles in the Poincare recurrence map, resulting in what seemed to be an ergodic dynamics from an irrational rotation number map on a torus. To have a global representation of this dynamics, we calculated the angle between the axial direction and the average velocity field, and compared it to the migration angle calculated from the pseudo-periodicity. We define the average velocity as

$$\langle \vec{U} \rangle = \frac{1}{A} \int_A \vec{U}(\vec{r}) \, dA$$

where $\vec{U}(\vec{r})$ is the velocity in position $\vec{r}$ of the unit cell and $A$ is the area of the unit cell. The angle $\theta$ between the average velocity and the axial direction is

$$\theta = \tan^{-1} \left( \frac{\langle \vec{U} \rangle_y}{\langle \vec{U} \rangle_x} \right)$$

where $\langle \vec{U} \rangle_x$ and $\langle \vec{U} \rangle_y$ are the x-component and y-component of the average velocity $\langle \vec{U} \rangle$.

Figure S1 shows the dependence of the single trajectory based migration angle (Fig. S1A, same as Fig 4A in the manuscript) and the average velocity angle (Fig. S1B) on the ratio between pillar radius $D_0$ and the pitch distance $D_y$ in y-direction. The two results, one using the recurrence map and individual trajectories, and the other computed using the velocity field directly, are remarkably close to each other. This suggests that the pseudo-periodicity described in the main text is a manifestation of the anisotropic permeability of the flow. Even when the average migration angle can be directly calculated from the average velocity in the unit cell, the Poincare recurrence map
introduced in our paper is essential to elucidate individual trajectories, and the structure of the flow lanes. In addition, the recurrence map allows us to calculate the finite-radius particle trajectories resulting from the interplay between the flow of individual particles and particle-pillar interaction.

![Figure S1](image.png)

**Figure S1** Normalized migration angle of particle trajectories and average flow velocity. (A) The migration angle obtained using the Poincare recurrence map and normalized by the pillar structural angle. \( \theta_N \) is the migration angle calculated from a pseudo-periodicity \( N \) and \( \theta_p \) is the structural pillar array angle determined by \( \tan^{-1}\left(\frac{D_x}{d_p N}\right) \). (B) The angle of the average flow velocity in a unit cell normalized with the structural pillar array angle. \( D_x, D_y, D_0 \) are the pitch in \( x \) and \( y \) direction and the pillar diameter.

**Recurrence map in a point-like pillar array**

In general, the analytical form of the recurrence map \( f \) is unknown and needs to be computed using Stoke’s equation with relatively complex boundary conditions. However, in the limiting case of negligible pillar diameter (point-like pillar system) the flow is not perturbed by the pillar array so that particles’ trajectories are parallel to the \( x \)-axis of the array. In the recurrence-map coordinates, if a particle was at position \( \eta_i = (y_i/D_y) \) at the inlet of the unit cell, the position of the particle \( \eta_{i+1} = (y_{i+1}/D_y) \) at the next pillar inlet becomes \( \eta_i - \epsilon/D_y \) because in the unit cell the \( y \) coordinates are measured with respect
to the bottom pillars, and the next pillar position has a y-axis shift by $\epsilon$. (Note that in the main text we defined $\eta$ as $y/G$ but in a point-like pillar array $G = D_y$, and $\eta = y/D_y$.) Thus, in this case the recurrence map can be calculated analytically as following.

$$\eta_{i+1} = f(\eta_i) = \begin{cases} 1 + \eta_i - \epsilon/D_y & \text{if } \eta_i < \epsilon/D_y \\ \eta_i - \epsilon/D_y & \text{if } \eta_i > \epsilon/D_y \end{cases}$$

where we have used the unit cell periodic boundary conditions.

From the equation above we can calculate the local periodicity and the migration angle. Assume that the pillar array has a structural periodicity of $N_p$. This means that the array repeats its structure for translations of the form $x + N_pD_x$ in the x-direction. This symmetry requires that the row shift in the y-direction be $\epsilon = D_y/N_p$. We can calculate the sequence of positions measured at the inlet of the unit cell as the particle goes downstream. By iterating the recurrence map

$$f^{N_p}(\eta_0) = f\left(f^{N_p-1}(\eta_0)\right) = \eta_0 - N_p\frac{\epsilon}{D_y} = \eta_0 - 1 \equiv \eta_0$$

we see that the map returns to the original coordinate $\eta_0$ after $N_p$ iterations. (At the last step in the previous sequence of iterations we used the periodic condition of map in the y-direction.) Therefore, the initial position $\eta_0$ is repeated over every $N_p$ iterations. The sequence of these positions are:

$$\{\eta_0, \eta_1, ..., \eta_{N_p-1}, \eta_0\}.$$

In this sequence, there are $N_p - 1$ direct transitions and one veering transitions. Because the flow periodicity and the pillar structural periodicity is same, the migration angle of small particle trajectories is zero. The lane dividing separatrices happen at inlet coordinates $\epsilon, 2\epsilon, ..., (N_p - 1)\epsilon$. Particles starting at those points will end in stagnation points. Trajectories of particles within the flow lanes always have a periodicity of $N_p$.

We can also use the recurrence map to study multi-directional sorting modes. For the case of $\epsilon = MD_y/N$ where $M, N$ are co-primes, the recurrence map $f$ is

$$f(\eta) = \begin{cases} \eta - \frac{M}{N} & \text{if } \eta > M/N \\ \eta - \frac{M}{N} + 1 & \text{if } \eta < M/N \end{cases}$$

With this recurrence map, we can calculate the $N$-th iteration position:

$$f^N(\eta) = \eta - M \equiv \eta.$$

In the last step, we use the periodic boundary conditions and the fact that $M$ is an
integer number. Therefore, the initial position is repeated every $N$ steps. However, in this case, the particle goes to the adjacent unit cell $M$ times before it lands in the same original position $\eta$. Thus, the pseudo-periodicity in this case is $N/M$. Note that in this geometry, the symmetry of the streamlines still coincides with the symmetry of the pillar structure.

**Local periodicity**

The local periodicity of a long trajectory shows an intermittent behavior. Fig. S2 A–C shows several cycles in a long trajectory. Depending on the initial position, these cycles have local periodicities of any of two consecutive integer numbers $N'$ and $N' + 1$. The order in which $N'$ or $N' + 1$ occur is irregular. Still we can define the pseudo-periodicity by averaging over multiple local periodicities in a large trajectory. Thus, the average periodicity, which we call the pseudo-periodicity, is a non-integer number between $N'$ and $N' + 1$.

![Figure S2. Long particle trajectory in a pillar array and the corresponding recurrence map. (A) The first cycle trajectory from one zigzag transition to next zigzag transition with local periodicity $N'=10$. (B) The second cycle trajectory with local periodicity $N'=11$. (C) The third cycle trajectory with local periodicity $N'=10$. (D) The recurrence map with $N'=10$ corresponding to the trajectory in (A). (E) The recurrence map with the next $N'=11$ particle positions overlaid with the previous cycle. (F) The recurrence map with...](image-url)
Not every initial position at the inlet will have an infinite trajectory: some positions will, upon a finite number end up in the stagnation point at some pillar downstream. We call that set of points the stagnation set. To find the stagnation set, we first identify the inlet point \( \eta_s \) where the recurrence map is discontinuous \( f(\eta_s^-) = 1; f(\eta_s^+) = 0 \). This is the starting point of the streamline that divides the veering streamlines \( \eta < \eta_s \) from the direct streamlines \( \eta > \eta_s \). The stagnation set can be computed by iteratively applying the inverse of the recurrence map \( f^{-1} \) to find all the points that the map to \( \eta_s \).

\[
\{ \eta_0 = \eta_s \rightarrow \eta_{-1} = f^{-1}(\eta_0) \rightarrow \eta_{-2} = f^{-1}(\eta_{-1}) \ldots \rightarrow \eta_{-l-1} = f^{-1}(\eta_{-l}) \rightarrow \cdots \}
\]

In general, this set is infinite. However, in the point-like pillar array, \( \eta_s = \frac{\epsilon}{D_y} = \frac{1}{N} \), the above set is finite:

\[
\left\{ \frac{1}{N}, \frac{2}{N}, \ldots, \frac{N-1}{N}, 1 \right\}.
\]

Note that the fluid flow brings the first position in this set, \( \eta_0 = 1/N \), towards the stagnation point on the next pillar after one iteration. The point \( \eta_{-1} = 2/N \) ends on the stagnation point after two iterations, and so on.

The pseudo-periodicity can be defined by the following formula:

\[
\bar{N} = \lim_{L \to \infty} \frac{1}{L} \sum_{i=1}^{L} N_i^L
\]

where \( L \) is the number of cycles in a long trajectory and \( N_i^L \) is local periodicity in the \( i \)-th cycle. In practice, the trajectory starts at some point \( \eta \) in the inlet of the unit cell. If is point belongs to the stagnation set, these iterations will eventually land on a stagnation point, and the computation of the pseudo-periodicity could proceed by iterating the map backwards. In any case, the stagnation set is countable (we can in principle count the pillars where the stagnation points will be) and therefore, this is a set of null measure. Therefore, with probability 1 the initial position \( \eta \) will not be in the stagnation set and we can find the next transition point infinitely.

**Geometric dependence of the recurrence map**

The recurrence map \( f \) is the mapping from the initial position at the inlet to the final position at the outlet. Therefore, as the streamlines veer differently depending on the geometry of the array, the recurrence map changes accordingly. Figure S2 shows the recurrence maps corresponding to different geometries, specifically differences in pillar
diameter. As the pillar diameter increases, the mapping $f$ approaches the identity function $y = x$ and the local periodicity increases considerably. In this case the dynamics of the iterations resemble that of a dynamical system after undergoing a tangent bifurcation, which has been described as a route to intermittent chaotic behavior. In Fig. S2 the initial position was set at the middle of the gap and the map was iterated until the trajectories went through 400 cycles.

As the pillar diameter increases, the recurrence map approaches a tangent bifurcation due to two factors. One factor is the discontinuity points moves closer to the pillar surface, and the bundle of veering streamlines gets compressed. The second reason is that the curvature of a recurrence map near the discontinuity point increases, indicating that the streamlines close to the pillars are considerably perturbed. Thus, the ratio between the pillar diameter and the pillar to pillar distance strongly determines the shape of the recurrence map.

The transition cycles on the recurrence map has a rich variety of patterns. Fig. S3 shows two such patterns. One is the case in which the local periodicity persists but the final position of a cycle does not match the initial position of a cycle exactly, and there is a smooth drift of the subsequent cycles making the trajectories to form the band shown in Fig. S3 (A), (C). The other case is when the local periodicity oscillates between $N'$ and
$N' + 1$. In this case the starting points of subsequent cycles may differ sufficiently that the local periodicity of subsequent cycles is different. Fig. S3 (B) and (D) shows that case in which the gray band in the recurrence map is more space filling than in the other two examples.

**Reconstruction of trajectories**

We derived a formula to connect the migration angle to the average streamline periodicity and the pillar structural periodicity.

$$\tan \theta = \frac{(\bar{N} - N_p)D_y}{\bar{N}N_pD_x}$$

where $N_p$ is the pillar structural periodicity and $\bar{N}$ is the streamline periodicity. To confirm the calculation, we computed the particle trajectories over multiple pillar arrays using the calculated recurrence maps, as shown in Figure S4. The particle trajectories with three different initial positions are shown as green lines with zigzag type transitions in red. As discussed in the text, as the pillar diameter $D_0$ increases, the length of one cycle increases regardless of initial position (Fig. S4 A–D). Given the same initial positions, we can observe the migration angle increase as the pseudo-periodicity increases.
We can also check the lateral spreading from the three different trajectories. As the particles move, the lateral spread in the y-axis oscillates but then converges back to the original spacing after a single cycle. Therefore, in the deterministic case the pillars do not spread out the particle trajectory in the y-direction.

**Initial position dependence on pseudo-periodicity**

In a point-like pillar array, every trajectory is periodic and therefore all cycles have local periodicity $N_p$. In arrays with pillars of finite size, however, the pseudo-periodicity estimated as

$$
\bar{N} = \frac{1}{L} \sum_{i=1}^{L} N_i'
$$

of two trajectories starting at different initial positions are not always identical due to the finiteness of computation. Here, $L \gg 1$ is the number of cycles over a long trajectory.

Figure S5 shows a typical plot of the pseudo-periodicity as a function of the initial position of the long trajectory. We observed that most values are around the averaged
pseudo-periodicity ($\langle N \rangle = 10.81$) over all initial positions. The local periodicity alternates irregularly between 10 and 11. That the averaged periodicity $\langle N \rangle = 10.81$ means the local periodicity of 11 occurs more often than that of 10 over a long trajectory. In the experiments performed for this paper the particles were injected in the whole inlet of the array, and therefore the migration angle was estimated from the collective average over all individual trajectories. Therefore, our definition of averaged pseudo-periodicity is a reasonable choice to characterize the migration angle.

![Figure S5. Pseudo-periodicity dependence on the initial position. Here, pillar diameter is 80nm and $D_x = D_y = 400\, \text{nm}$. The pillar has a structural periodicity of $N_p = 10$.](image)

**Recurrence map using 2 cells**

Our simulations assumed a pillar array in a periodic lattice, which allowed us to solve the Stoke's equation in the unit cell. While this is a good approximation to model the actual system, this assumption may impose some limitations on the interpretation of the actual trajectories measured in finite size arrays in terms of our model. Here we will study two possible ways in which this approximation may influence our conclusions: one is the validity of our assumed pressure boundary conditions at the inlet and outlet of a unit cell, the other is the effect of the wall boundaries of the microchannel housing the pillar array on the particle trajectories. These two assumptions may be the cause of the differences between model and experiments seen in Figure 4 of the main.

The use of a single unit cell implies an infinite array in the x, y-directions. However, the pressure and velocity distributions of a multi cell array in which the boundary conditions are imposed at the input and output cells may be different from the pressure and velocity distributions if the boundary condition are imposed at the single unit cell. In our simulations, we applied a constant pressure on the inlet and outlet (AH, DE in Figure 1). To test that this is not too severe an approximation, we simulated several set
of combined unit cells (2x1, 2x2, 3x3 unit cells, still with periodic boundary conditions) to verify consistency with the 1x1 unit cell simulations. The resulting recurrence map showed no significant difference in either case.

The boundary conditions at the microchannel housing the pillar array may also have an impact on the fluid flow. In our experiments, the use of periodic boundary condition to mimic the infinite array can be justified by the fact that our nanoDLD device has 125x1250 unit cells inside the 50 µm wide microchannel. Thus, we can justify that our assumption of an infinite array is valid in the bulk of our arrays, as long as we are far from the boundaries. This is further justified by the observation that the experimentally measured migration angle matches well the one estimated with our model. To be more quantitative we simulated multiple arrays (15x20, 30x20, 45x20, 60x20 arrays) and observed that the effects of the walls started to be washed off at ~20 unit cells away from the microchannel boundary. The effect of the wall boundary conditions on particle trajectories in DLD arrays has also been discussed in 8.

**Movie Description**

The concept of the recurrence map can be more clearly understood using an animated description of the dynamics (Movie bump.mp4). Our animation shows the time evolution of a particle trajectory and connects it to the corresponding dynamics in a recurrence map. In the movie, the three panels from the left show the trajectory in multiple pillars, the trajectory in a unit cell and the recurrence map, respectively. Here, \( N_p = 5 \) and the pillar diameter \( D_0 \) is 200 nm. The pillar to pillar distance \( (D_x = D_y) \) is 400 nm so that the ratio \( (D_0/D_y) \) is 0.5. The long trajectory is composed of total 8 transition segments starting from 90\% of the pillar gap \( (\eta_0 = 0.9) \).

For the particle with radius larger than the critical diameter \( (D_c) \), the trajectory is similar to that of the small particle until it veers around the pillar for the first time. But due to the shift of the center of particle by the pillar repulsion, it cannot veer around the pillar and bumps on the post surface. After it follows the closest possible streamline to the separatrix line, it ends up to the same initial position (the directional locking 9). The animation in the middle shows this transition by overlaid trajectories inside a unit cell. In the recurrence map, the mapping \( f \) intersects the identity function \( (y = x) \) which becomes a fixed point in the dynamics. Here, blue circle means the initial position at the inlet and the red circle means the final position at the outlet.

**Geometric parameters in the unit cell simulation**

Four key parameters are used to specify the geometric configurations in our arrays, as shown in Fig. 1A: pillar diameter, \( D_0 \), the lattice parameters (pitch) of the pillar unit cell, \( D_x, D_y \) and the row shift fraction, \( \epsilon \). The value of these geometric parameters in the pillars used in this paper are summarized in the following Table S1.
Here, we fixed the array structural periodicity as \( N_p = 10 \) and the pitch distance in y-direction at 400nm \( (D_y = 400 \, \text{nm}) \). Therefore, the only independent variables are the pillar diameter \( D_0 \) and the pitch distance in x-direction \( D_x \). However, we also tested the different structural pillar periodicities \( (N_p = 3, 4, 5, 10) \) in different simulations.

### References


