Orientational and directional locking of colloidal clusters driven across periodic surfaces

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When particles are driven across crystalline surfaces, their trajectories do not necessarily follow the applied force but become locked to the substrate lattice directions. Such directional locking, being relevant for bottom-up nano-device assembly [1,2] and particle sorting [3-6] has been intensively studied for isolated or single particles [3-11]. Here we experimentally study the motion of extended colloidal clusters sliding over a periodically corrugated surface. We observe that both their orientational and center-of-mass motions become locked into directions not coinciding with the substrate symmetry but determined by the geometrical moiré superstructure formed by the cluster and substrate lattices. In general such moiré superstructures are not strictly periodic which leads to competing locking directions depending on cluster size. Remarkably, we uncover a dependence of directional locking on higher Fourier components of the surface corrugation profile which can be tuned on atomic surfaces via the external load [12,13]. This allows for an unprecedented control of cluster steering relevant for nano-manipulations on surfaces.

An important step towards the bottom-up assembly of nanoscopic functional components from atomic building blocks is the controlled translation and positioning of atoms and molecular clusters on surfaces by external forces [14]. At macroscopic scales, objects typically follow the direction of an external force. This is no longer true for microscopic components on atomically corrugated substrates, where particle trajectories can lock to substrate lattice directions since they provide low energy corridors within the potential energy landscape. Such directional locking has been previously observed for nanocrystals migrating over atomic surfaces [1,2] and microparticles on arrays of obstacles [3-5] or optical traps [6,7] but also in flux flow of type-II superconductors [8]. In addition to their fundamental understanding, deviations between the direction of particle motion and the applied force have important consequences for the manipulation of atoms on surfaces and must therefore be considered in bottom-up assembly strategies. Contrasting with isolated or weakly interacting particles [8-11], however, little is known about the driven motion of spatially extended crystalline clusters across periodic surfaces [15]. Here we experimentally and numerically study the translational and the orientational motion of micron-sized colloidal clusters with up to $N = 400$ particles which are driven across a patterned surface. Although the governing forces and their range is very different in colloidal systems, tribological experiments suggest a close resemblance with observations in atomic systems [16]. Such system-independent features are in perfect agreement with the Frenkel-Kontorova model [17] which ignores all details regarding the relevant forces and just considers a monolayer of interacting particles on a corrugated surface. Compared to atomic clusters, where well-defined driving forces are difficult to realize by means of scanning probes [18,19], colloidal clusters can be

precisely and continuously driven by electric [20], magnetic [21] or gravitational [22] fields across substrates. With typical particle sizes of the order of microns, colloidal particles' positions can be optically determined with sub-percent resolution (compared with their diameter), which allows to precisely resolve their motion relative to the substrate [16,23,24].

We demonstrate the occurrence of nontrivial and stable locking directions accompanied with specific locking orientations, which mainly depend on the lattice mismatch and cluster size. Such parameters offer the opportunity to control the microscopic dynamics, and to select specific locking directions. Via an analytical expression for the average particle potential energy, we reveal a surprising relationship between the Fourier components of the particle-surface interaction profile and the specific locking phenomenology, which has also immediate practical consequences for the nano-manipulation of objects.

Substrates with hexagonally-arranged circular dimples or wells (lattice spacing b = 5.8 μm, diameter 4.2 μm, and depth 80 nm) were created by photolithography. Colloidal clusters were made from an aqueous suspension of colloidal particles (diameter a = 4.45 μm) where a small amount of polyacrylamide (PAAm) was added. Because the colloids contain iron-oxide inclusions, PAAm causes strong interparticle bridging flocculation [25,26] which results in rigid 2D clusters with a lattice constant a (Fig.1a) and broad distribution in cluster size and shape. These clusters are driven across the substrate with a constant gravitational force $F = mg \sin\alpha$ acting on each particle by tilting the entire setup by an angle α (Fig.1b). The microscope stage can be also rotated along its perpendicular axis so that the direction of **F** in the *x*-*y* plane can be varied. The positions of colloidal particles relative to substrate wells are determined by video microscopy. (More details are found in the Methods section.)

Fig.1c shows the initial and final configurations of a sliding cluster, connected by the center-of-mass trajectory (green). Clearly, its direction of motion $\theta_d = 19.1°$ is different from the direction of the driving force $\varphi_F = 2.9°$, both measured relative to the substrate orientation *x*. It should be noted that, in addition to θ_d, also the cluster's orientation θ_o is preserved during the sliding. For the particular example shown here, $\theta_o = \theta_d = 19.1°$ due to the specific choice of a/b (an example where $\theta_d \neq \theta_o$ is shown in Fig.S1). Fig.1d shows the value of θ_d and θ_o for two differently-sized clusters (N = 86 and N = 46 particles) as a function of the driving direction φ_F. For both clusters, two broad plateaus in θ_d are observed at 19.1° and 79.1°, indicating strong directional locking. At each plateau, the orientation θ_o also remains locked near 19.1°. In the angular range between the two plateaus, the cluster motion follows closely the direction of the driving force, while its orientation fluctuates irregularly. Molecular-dynamics simulations results (guided by solid curves in Fig.1de) reproduce the experimental trends (Methods). Given the geometric conditions, directional locking is remarkably robust over cluster size and shape, as shown in Movie 1 of the Supplementary Information (SI).

Fig.1: Observation of orientational and directional locking. a,b, Schematic illustration of a rigid cluster formed by bridging flocculation, and driven by the gravitational force $F = mg \sin\alpha$ across a tilted corrugated substrate. **c,** An experimental image showing the locked moving direction of a cluster (a = 4.45 μm) on top of a patterned surface (b = 5.80 μm). The cluster's center of mass trajectory is shown in green. The cluster orientation θ_o, the direction of motion θ_d and the driving force angle φ_F are defined relative to the substrate principal direction *x.* Scale bar: 30 μm. **d,** The directional and orientational locking effects for two different clusters with N = 86 and N = 46 particles on top of structures obtained at various driving force orientations φ_F and magnitudes F = 62.9 fN (α = 12.7°) and F = 71.6 fN (α = 14.5°) respectively. Full symbols stand for directions, open symbols stand for orientations. Semi-transparent data points with

connecting lines correspond to simulation data. Each experimental data point is obtained from a cluster trajectory (examples in insets) of about 20 μm to 30 μm in length, see Movie 2 and Movie 3 of the SI. As reference, the dashed line $\theta_d = \varphi_F$ denotes the direction of motion, parallel to the external driving, approximately followed by the clusters between the two plateaus.

Fig.2a shows the translational and orientational motion of the center of mass of a cluster driven on the same substrate at $\varphi_F = 0°$ and $F = 71.6$ fN. The periodic stair-like behavior of the *x(t)* and *y(t)* curves indicate a stick-slip-like motion over the tilted periodic potential energy landscape. The cluster exhibits directional locking at $\theta_d \approx 19.1°$ as long as its orientation remains (for the specific choice of a/b as discussed here) near $\theta_o \approx 19.1°$. As soon as the cluster rotates away by ~1° (indicated by arrows), directional locking is lost. This behavior originates from the geometrical superstructure (moiré pattern) resulting from the nearly perfect matching of the θ_o-rotated colloidal lattice with the substrate pattern. Fig.2b shows a time series of a sliding cluster where each particle has been color-coded according to its distance Δr_{min} to the nearest substrate potential well. The moiré superstructure becomes clearly visible by those particles which are almost perfectly trapped at substrate minima, namely having a very small displacement Δr_{min} (blue particles in Fig.2b at $t = 79$ s and $t = 88$ s). Such superstructure repeats periodically in time, with each cycle corresponding to a stick-slip event (Movie 4). To better illustrate the superstructure, in Fig.2c we draw an incommensurate hexagonal cluster on top of the periodic substrate. In general, ideal, or strictly periodic, moiré superstructures between two lattices occur whenever a set of their lattice points coincide, namely

$$n_1 \mathbf{b}_1 + n_2 \mathbf{b}_2 = m_1 \mathbf{a}_1 + m_2 \mathbf{a}_2, \qquad (1)$$

where $\mathbf{a}_i$ and $\mathbf{b}_i$ (i = 1, 2) are the corresponding lattice vectors and n_i and m_i integers. For triangular lattices, this is achieved when the ratio between the cluster periodicity $a = |\mathbf{a}_1| = |\mathbf{a}_2|$ and the substrate periodicity $b = |\mathbf{b}_1| = |\mathbf{b}_2|$ fulfills

$$a/b = \lambda, \qquad (2)$$

where $\lambda = ((n_1+n_2/2)^2 + 3n_2^2/4)^{½} / ((m_1+m_2/2)^2 + 3m_2^2/4))^{½}$. From equation (1) one obtains

$$\theta_o = \theta_s + \arctan(\sqrt{3}/2\, m_2 / (m_1+m_2/2)), \qquad (3)$$

where $\theta_s = \arctan(\sqrt{3}/2\, n_2 / (n_1+n_2/2))$ is the orientation of the superstructure lattice vector $\mathbf{s} = n_1 \mathbf{b}_1 + n_2 \mathbf{b}_2$. Experimentally, even though the lattice-spacing ratio a/b always deviates marginally from equation (2), a locally-periodic moiré superstructure (illustrated in Fig.2c) will form and promote directional locking. For a/b = 4.45/5.80, the observed moiré superstructure (Fig.2b,c) is (n_1, n_2, m_1, m_2) = (2, 0, 2, 1), which leads to $\theta_o = 19.1°$ according to equation (3), in excellent agreement with our experimental results. Other choices of (n_1, n_2, m_1, m_2), which also approximately satisfy equation (2) but are not observed in experiments, are discussed in SI, Table S1.

To understand the stick-slip dynamics and the direction of motion from an energetic point of view, in Fig.2d we show the calculated potential energy landscape (Methods) of a rigid cluster (inset Fig.2a) with fixed orientation $\theta_o = 19.1°$. Superimposed as green points, we plotted the experimentally observed trajectory of the cluster from $t = 0$ s to $t = 34$ s. Clearly, the cluster follows a corridor of lowest energy lying close to the direction of the driving force. The superstructure configurations in Fig.2b correspond to the minima of the potential energy for the cluster. The stick events occur whenever the cluster climbs the inter-minima barriers. The energy corridors are a result of the average potential per particle, or "lock-in potential"

$$U(\Delta \boldsymbol{r}, \theta_o) = \sum_{\boldsymbol{Q}} \tilde{V}(\boldsymbol{Q}) \cos(\boldsymbol{Q}\cdot\Delta\boldsymbol{r}) \qquad (4)$$

which is evaluated in the Methods section for a rigid cluster rotated by θ_o relative to the substrate symmetry direction and translated by $\Delta\boldsymbol{r}$. Here $\tilde{V}(\boldsymbol{Q})$ are the Fourier components of the single-particle potential $V(\boldsymbol{r})$ evaluated at the reciprocal lattice vectors $\boldsymbol{Q}$ of the colloidal crystal. For an infinite colloidal crystal, $\tilde{V}(\boldsymbol{Q})$ is nonzero only when $\boldsymbol{Q}$ equals a reciprocal vector of the

substrate lattice $\boldsymbol{G}$, which occurs only when equation (1) is satisfied exactly. The leading contribution to $U(\Delta \boldsymbol{r}, \theta_o)$ results from cosine functions of the set of the shortest common reciprocal vectors: this determines the direction of the low-energy corridor on the energy landscape at $\theta_d = \theta_o - \theta_s$, and creates a periodicity (dashed line of Fig.2d) significantly shorter than the discrete lattice spacing of the substrate (solid arrow in Fig.2d). (See Methods for more details.) Most remarkably, according to equation (4), a periodic potential composed of only the first-order Fourier components, namely a superposition of single-frequency sinusoidal potentials being often assumed in simplified tribological models, would not generate directional locking at all (see Fig.S2). To intuitively appreciate the importance of the higher Fourier components, consider the displacements of the colloid particles relative to the substrate minima. For an ideal moiré superstructure, any particle in the cluster is translationally equivalent to one within its unit cell (for example one of the 7 particles within the dashed hexagon of Fig.2c). The unique displacements of those few particles induce fine structures within the primitive cell which can only couple to the higher Fourier modes of the substrate potential. These observations hold exactly for infinite lattices with equation (1) perfectly satisfied. For finite cluster sizes, however, a neighborhood $\boldsymbol{Q} \approx \boldsymbol{G}$ also contributes to $U(\Delta \boldsymbol{r}, \theta_o)$, thus directional locking can be observed with imperfect matching.

Fig.2: Moiré pattern and energy landscape on b = 5.80 μm substrate. a, Translational displacement $x(t)$, $y(t)$, orientation $\theta_o(t)$ and velocity direction $\theta_d(t)$ of a cluster (inset) under drive F = 71.6 fN at $\varphi_F = 0°$. **b,** A sequence of color coded images at t = 79 s, 82 s, 85 s, 88 s during the motion of the cluster. Colors represent the distance of the particle to the center of a nearest well Δr_{min}. **c,** A cluster with primitive vectors $|\mathbf{a}_1| = |\mathbf{a}_2| = a = 4.45$ μm and orientation $\theta_o = 19.1°$ on top of a substrate with primitive vectors $|\mathbf{b}_1| = |\mathbf{b}_2| = b = 5.80$ μm. Blue particles denote those closest to substrate minima. They form a locally-periodic superstructure. The triangle rule denotes $2\mathbf{b}_1 \approx 2\mathbf{a}_1 + \mathbf{a}_2$. The 7 particles within the dashed hexagon form a unit cell of the superstructure. **d,** The per-particle potential-energy $U(\Delta \boldsymbol{r}, \theta_o = 19.1°)$ landscape of the cluster in (**a**). Green data points show cluster positions at 0.33 s intervals.

Another important consequence of imperfect matching, or locally-periodic moiré superstructures, is that directional locking becomes dependent on the cluster size. This effect becomes evident by considering a different substrate periodicity, b = 5.40 μm, which allows us to observe completely different locking directions and to investigate two competing moiré superstructures. As shown in Fig.3a, small clusters ($N < 40$) prefer the $(n'_1, n'_2, m'_1, m'_2) = (1, 1, 2, 0)$ moiré with $\theta'_s = 30.0°$, $\theta'_o = 30.0°$, $\theta'_d = 0°$ while large clusters ($N > 90$) prefer $(n''_1, n''_2, m''_1, m''_2) = (3, 0, 3, 1)$ with $\theta''_s = 0°$, $\theta''_o = 13.9°$, $\theta_d = 13.9°$ (Movie 5). The colored patterns of Fig. 3a insets highlight the stable superstructures in the two regimes. Both superstructures are observed when $40 < N < 90$: a cluster can rotate from one superstructure to the other during sliding (Movie 6). These results are complemented by the numerical simulations in Fig.S3. The reason behind this size effect is the competition between the superstructure periodicity $s = |\mathbf{s}| = |n_1 \mathbf{b}_1 + n_2 \mathbf{b}_2| = b\,((n_1+n_2/2)^2 + 3n_2^2/4)^{½}$ and the imperfectness $\delta = 1/\lambda\, a/b - 1$ of the ratio a/b, which is illustrated in Fig.3b,c. According to equation (2), for a strictly periodic superstructure, δ should vanish. The (1, 1, 2, 0) moiré has a smaller s' = 9.35 μm but larger δ' = -4.8% (Fig.3b), compared with the (3, 0, 3, 1) moiré which has s'' = 16.20 μm but a better match δ'' = -1.0% (Fig.3c). Small clusters (shaded region in Fig.3b) are not affected by the larger δ' and therefore favor the smaller s' with denser trapped particles, while large clusters are much more sensitive to the imperfect commensurability and therefore would trade the number of imperfectly trapped particles (Fig.3b) for a smaller δ'' (Fig.3c) to minimise the total energy. Fig.3d shows the

calculated minimum potential energy E as a function of cluster size N (Methods) for clusters in both superstructures. These calculations confirm that clusters with $N \gtrsim 100$ have lower energy in the $\theta_o = 13.9°$ superstructure while for smaller clusters the two superstructures compete. Note that Fig.3a represents 24 different clusters with different and irregular shapes in experiments. Fig.S4 reports a discussion of the influence of cluster shape.

Fig.3: Dependence on the cluster size on b = 5.40 μm substrate. **a,** The observed stable orientation θ_o of clusters and the corresponding stable direction of motion θ_d as a function of cluster size for a substrate. We classify θ_o and θ_d as stable if the cluster has translated by 5 lattice constants without noticeable changes in the angles. The two main stable orientations observed in experiments (open symbols) are $\theta_o \approx 30°$ and $\theta_o \approx 13.9°$ (dotted lines), the corresponding superstructures (insets) are $(n'_1, n'_2, m'_1, m'_2) = (1, 1, 2, 0)$ and $(n''_1, n''_2, m''_1, m''_2) = (3, 0, 3, 1)$. **b, c**, Illustration of the locally-periodic superstructures formed by trapped particles (blue) at $\theta_o \approx 30°$ and $\theta_o \approx 13.9°$ respectively for a symmetric cluster of 61 particles. The triangle rule in **b** denotes $\mathbf{b}_1 + \mathbf{b}_2 \approx 2\mathbf{a}_1$ and in **c** denotes $3\mathbf{b}_1 \approx 3\mathbf{a}_1 + \mathbf{a}_2$. The blue particles are badly trapped near the edge of the cluster in **b** due to its large lattice mismatch. **d,** The mean potential energy per particle as a function of cluster size N for clusters of different shape in the two different superstructures $\theta_o = 30°$ and $\theta_o = 13.9°$. The dashed line indicates the potential energy in the $N \to \infty$ limit for incommensurate rigid lattices.

Directional locking is also influenced by the strength of F. Fig.4a, shows the experimentally measured cluster velocity for three different driving directions φ_F. Clearly, they were locked to $\theta_d = 19.1°$ only at low driving forces but directional locking becomes lost when the driving force exceeds a certain value F_c. Here the measured F_c in experiments is 89 fN and 106 fN for $\varphi_F = 0.08°$ and 4.23° respectively. The difference arises from the different deviation of driving direction to the low-energy corridor shown in Fig.2d. To escape this corridor, it takes a critical perpendicular force $F_{c\perp} = F_c \sin(|\theta_d - \varphi_F|)$. Given the above experimental results, we obtain $F_{c\perp} \approx 28$ fN. Fig.4b shows the sliding direction θ_d (color coding) in the F_x-F_y plane obtained via zero-temperature simulations. Two ribbon-like regions with uniform values of $\theta_d = 19.1°$and $\theta_d = 79.1°$ are observed near $\varphi_F = 19.1°$ and $\varphi_F = 79.1°$ respectively, corresponding to the directional locking. The ribbon-like regions confirm the above picture of $F_{c\perp}$. Indeed, via nudged elastic band simulations [27], we have evaluated $F_{c\perp} \approx 30$ fN, which agrees with the experimental estimation.

Fig.4: Influence of driving force. a, The sliding direction θ_d as a function of the driving force F for three different driving directions $\varphi_F = 0.08°$, 4.23° and 9.74° for three clusters, of 98, 116 and 57 particles respectively. The measured $F_{c\perp}$ is 29 fN and 27 fN for $\varphi_F = 0.08°$ and 4.23° respectively. Given $F_{c\perp} \approx 28$ fN, the critical force for $\varphi_F = 9.74°$ would be $F_c \approx 172$ fN, exceeding the maximum accessible force of 138 fN. **b,** The simulated sliding direction θ_d (color coded) as a function of the driving-force components for a 61-particle hexagonal cluster sliding across the b = 5.80 μm substrate at $T = 0$ K. The dark region near the origin is statically pinned. Directional locking is highlighted by the constant-θ_d rectangular bands. The 3 symbols represent the experimental maximum force in the locked regime (from panel **a**) in the corresponding direction.

The translational and orientational motion of driven clusters on patterned surfaces as reported here, is essentially the result of the competing symmetries between the two rigid lattices in physical contact. Down to the atomistic scale, such findings hit on the paramount playground of incommensurate 2D material heterojunctions [28-30], where, due to the extremely large in-plane stiffness, interface geometry rules the coupling between orientational and translational degrees of freedom via specific moiré reconstructions. Comparable observations are expected also in systems characterized by significant elasticity of the nanocontacting objects. An important consequence of equation (4) is that directional locking results from the higher Fourier components of the corrugation potential. Since overlayer-substrate interactions are typically distance-dependent [12,13], these higher Fourier components, and thus directional locking will depend on the load. The tuning of this normal load applied to the translated object opens a means of exploitation in nano-manipulation experiments. Future directions of research might consider the effect of grain boundaries, alternating different periodicities and/or orientation of the substrate pattern, which could provide further means of controlling directionally locked trajectories.

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Data availability

The data that support the findings of this study are available from the corresponding author upon request.

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Author contributions
C.B. and X.C. designed the experiments; X.C. carried out the experiments; A.V., N.M and E.P. wrote the computer code; E.P. performed the numerical simulations; X.C. and E.P. analysed the data. All authors contributed to the theoretical understanding, discussed the results, and wrote the paper.

Competing interests
The authors declare no competing interest.

Methods

Substrate preparation and characterization.
The periodic structure on the sample substrate was created by photolithography. A thin layer (~80 nm) of photoresist (a mixture of SU8 2000.5 and SU8 thinner at a volume ratio of 1:2) was spin coated to a glass substrate at a speed of 3500 rpm for 30 seconds. After a soft baking of about 1 min on a hot plate at 95°C, the substrate was exposed by UV light under a pre-designed mask that contains a hexagonal array of circular regions that are not transparent to light. After exposure, the substrate was baked for 3 min at 95°C before it was rinsed in SU8 developer for 40 seconds and in isopropanol for 5 seconds, after which the circular regions of the photoresist being blocked by the mask are dissolved away from the substrate, resulting in an array of wells on the glass surface. The SU8 structure was scanned under an atomic force microscope (AFM) with a Bruker OTESPA-R3 tip (tip curvature radius ~7 nm). The scans (Fig.S5) show that, for the b = 5.80 μm substrate, the diameter of the wells is 4.2 μm and the depth of the wells is 80 nm.

Sample preparation and cluster formation.
The colloidal suspension is composed of Dynabeads M450 dispersed in sodium dodecyl sulfate (SDS)-water solution at 50% critical micelle concentration. The Dynabeads are polystyrene particles that contain iron oxide inclusions. They have a diameter a = 4.45 μm, buoyant weight mg = 286 fN and a gravitational height of $h_g = k_B T/mg$ = 14.2 nm at room temperature T = 295 K. To realize the bridging flocculation effect, we add 0.02 percent of polyacrylamide (PAAm) by weight into the colloidal suspension. The large molecular weight M = 18,000,000 a.m.u. of the PAAm here greatly enhances the bridging flocculation effect [31]. The iron oxide inclusions in the dynabeads further selectively facilitate the bridging effect between the particles [25,26]. 0.01% of SDS by weight was further added to the colloidal suspension upon making the sample to reduce the sticking of particles to the substrate. The colloidal suspension is injected into a sample cell of about 20 mm × 30 mm × 300 μm in size, where 300 μm is the distance from the bottom substrate to the top cover slide. Under gravity, the particles sediment towards the bottom of the sample cell. Due to the bridging effect, any two particles getting in contact will tightly bind to each other within a few seconds. The initial particle coverage (~ 0.01) is so small that clusters can hardly grow larger than 10 particles. To facilitate the formation of larger clusters, the sample cell is tilted by 15~20 degrees so that clusters can translate under gravity. During their translation, clusters can collect particles on their way and grow larger (Movie 7). This process also clears up scattered isolated particles on the substrate so that they do not interfere with future cluster sliding. The typical cluster size in our experiments is N = 50~100 particles. The largest cluster we have obtained consists of 399 particles. When sliding on the structured substrate, the Péclet number [32] of a cluster is on the order of 10^2 or larger, namely the translational dynamics of the cluster is almost deterministic.

Stiffness of the clusters.
The clusters have very rigid structures and rarely change their shape during sliding. The rigidity of the clusters is characterized by the nearest-neighbour bond-length fluctuation $\Delta a_{ij} = a_{ij} - \langle a_{ij} \rangle_t$, where i, j are nearest neighbours and $\langle\rangle_t$ denotes an average over time. Fig.S6 shows the probability distribution $p(\Delta a)$ for all nearest neighbour bonds within a cluster, for two differently-sized clusters on flat surfaces. Considering the accuracy (10 nm) of particle positions, the upper bound of the bond length fluctuation is 9 nm only, less than 0.2% of the particle size. The formation of very stiff clusters is in contrast to much softer colloidal clusters which will form under the influence of a rotating magnetic field [33] or by DNA coating of the spheres [34].

Particle and cluster tracking, substrate characterization.
Experimental images were recorded at a 3 Hz frame rate. Using a standard particle-tracking algorithm [35], we can track the positions of the colloidal particles (accuracy 10 nm). To precisely identify particle positions relative to the substrate potential, we have to reconstruct the potential wells being optically obstructed by the colloidal particles. This is achieved by interpolation of the hexagonal structure of the lattice wells which is clearly visible around the cluster (Fig.S7). The orientation θ_o of a cluster is defined as the average nearest neighbour bond orientation along one of the symmetry direction (accuracy 0.01°). The direction θ_d is defined by $\theta_d = \arctan(v_y/v_x)$, where v_y and v_x are cluster center-of-mass velocity along y and x direction respectively. The accuracy of θ_d depends on the length of the trajectory and is typically 0.1°. In Fig.2a, $\theta_d(t)$ is calculated from a trajectory of 10 seconds, accordingly the accuracy in this case is 1°.

Modeling, molecular dynamics and energy calculations.
The substrate corrugation felt by a particle at position $\boldsymbol{r}$ is a sum of infinite terms $V(\boldsymbol{r}) = \sum_{n,m} V_{\text{dimple}}(|\boldsymbol{r} - (n\mathbf{b_1} + m\mathbf{b_2})|)$, where $\mathbf{b}_1$ and $\mathbf{b}_2$ are the primitive vectors of the substrate, $n,m \in N$. $V_{\text{dimple}}(|\boldsymbol{r}|)$ is the analytical potential energy profile of a sphere of radius R located at a distance $\boldsymbol{r}$ from the center of a cylindrical well with potential depth ϵ, linear depth h and width W. $V_{\text{dimple}}(\boldsymbol{r}) = -\epsilon$, for $|\boldsymbol{r}| < r_m$, $V_{\text{dimple}}(\boldsymbol{r}) = -\epsilon\,(1-\cos(\arcsin(-(|\boldsymbol{r}|-W)/R))\,R/h$, for $r_m < |\boldsymbol{r}| < W$, $V_{\text{dimple}}(\boldsymbol{r}) = 0$ for $|\boldsymbol{r}| > W$. The effective internal width of the well is $r_m = W - R\sin(\arccos(1 - h/R))$. To replicate the experimental results we use a potential depth ϵ = 105 zJ and widths W = 1.6 μm and 2.1 μm for substrates with periodicity b = 5.4 μm and 5.8 μm respectively. For the investigation of the clusters' dynamics we model the system as N particles interacting via LJ potentials, with the "truncated and shifted force" [36] cut-off at 1.6σ. The LJ parameters σ = 4.45 μm and ϵ_{LJ} = 7000 zJ are fitted to reproduce the experimental nearest-neighbour bond-length distribution at T = 295 K. The model includes no hydrodynamic interparticle interactions. We perform Langevin dynamics with a damping rate γ = 3.0 ms^{-1}, within a fourth-order Runge-Kutta integration scheme. To reproduce the per-particle potential-energy landscape as in Fig.2d we consider clusters of particles fixed at positions $\boldsymbol{R}_i = j_i\,\mathbf{a_1} + k_i\,\mathbf{a_2}$, where $\mathbf{a}_1$ and $\mathbf{a}_2$ are the primitive vectors of the colloidal lattice rotated at an angle θ_o, and the set $\{j_i, k_i\}$ for i = 1, 2, 3,..., N defines the cluster size and shape. The per-particle energy is then calculated as $U(\Delta\boldsymbol{r}, \theta_o) = 1/N \sum_i V(\boldsymbol{R}_i+\Delta\boldsymbol{r})$.

The lock-in potential and low-energy corridor.
As was done in Ref. [37] we evaluate the per-particle energy (or *lock-in potential* energy) of a lattice of N particles interacting with a periodic potential $V(\boldsymbol{r})$. All particles sit at positions $\boldsymbol{r}_j = \boldsymbol{R}_j + \Delta\boldsymbol{r}$, with $\boldsymbol{R}_j$ direct-space lattice points of the cluster. $\Delta\boldsymbol{r}$ is a rigid translation of the lattice relative to the origin of the axes. As independent variables we take the shift $\Delta\boldsymbol{r}$ and the mutual rotation θ_o of the particles lattice relative to the substrate potential. The per-particle interaction energy is

$$
\begin{aligned}
U(\Delta\boldsymbol{r}, \theta_o) &= 1/N \sum_j V(\boldsymbol{r}_j) && \text{(i)}\\
&= 1/N \sum_{\boldsymbol{R}} V(\boldsymbol{R} + \Delta\boldsymbol{r}) && \text{(ii)}\\
&= 1/N \sum_{\boldsymbol{R}} \sum_{\boldsymbol{G}} \tilde{V}(\boldsymbol{G})\, e^{-i\boldsymbol{G}\cdot(\boldsymbol{R} + \Delta\boldsymbol{r})} && \text{(iii)}\\
&= \sum_{\boldsymbol{G}} \tilde{V}(\boldsymbol{G})\, e^{-i\boldsymbol{G}\cdot\Delta\boldsymbol{r}} [1/N \sum_{\boldsymbol{R}} e^{-i\boldsymbol{G}\cdot\boldsymbol{R}}] && \text{(iv)}
\end{aligned}
$$

$= \sum_{\boldsymbol{G}} \tilde{V}(\boldsymbol{G})\, e^{-i\boldsymbol{G}\cdot\Delta\boldsymbol{r}} \sum_{\boldsymbol{Q}} \delta_{\boldsymbol{G},\boldsymbol{Q}}$ (v)

$= \sum_{\boldsymbol{Q}} \tilde{V}(\boldsymbol{Q}) \cos(\boldsymbol{Q}\cdot\Delta\boldsymbol{r})$. (vi)

Here $\boldsymbol{G}$, $\boldsymbol{Q}$ are reciprocal vectors of substrate and particles lattice respectively. $\tilde{V}(\boldsymbol{G}) = 1/\Omega \int_\Omega d\boldsymbol{r}\, V(\boldsymbol{r})\, e^{-i\boldsymbol{G}\cdot\boldsymbol{r}}$ is a Fourier coefficient of the substrate potential, where Ω is the area of its primitive cell. Finite N is initially considered and the limit $N \to \infty$ is performed at equation (iv). For finite N, as in our experiments, the Dirac deltas of equation (v) are to be replaced by finite-width peaks, that allow for nonzero $\boldsymbol{Q}$ to contribute to U, even if equation (1) is not satisfied exactly for any (n_1, n_2, m_1, m_2). Note that a superlattice primitive vector $\mathbf{s} = n_1\,\mathbf{b}_1 + n_2\,\mathbf{b}_2 = m_1\,\mathbf{a}_1 + m_2\,\mathbf{a}_2$ in real space gives rise to a superlattice primitive vector $\boldsymbol{Q}_s = -m_2\,\boldsymbol{\beta}_1 + m_1\,\boldsymbol{\beta}_2 = n_2\,\boldsymbol{\alpha}_1 - n_1\,\boldsymbol{\alpha}_2$ in reciprocal space, where $\boldsymbol{\beta}_1$, $\boldsymbol{\beta}_2$ and $\boldsymbol{\alpha}_1$, $\boldsymbol{\alpha}_2$ are reciprocal-space primitive vectors of the substrate lattice and particles lattice, respectively. $\boldsymbol{Q}_s$ is the shortest common reciprocal vector that provide the leading contribution to $U(\Delta\boldsymbol{r}, \theta_o)$. Given the triangular symmetry, there is a total of six equivalent shortest common reciprocal vectors. These mark three different directions: The direction parallel to $\boldsymbol{Q}_s$ and those at angles of ±120° from it. Each of the six reciprocal superlattice vectors creates a corrugation modulation in its own direction, and a "soft direction" perpendicular to that. The overall corrugation - with shorter periodicity $2\pi/(\sqrt{3}/2|\boldsymbol{Q}_s|)$ - is a superposition of these three corrugations, and is characterized by a low-energy corridor at an angle $\theta_d = \theta_o - \theta_s$. In addition to this general formalism, valid for arbitrary commensurability a/b, a simple geometrical approach to understand the direction of motion and reduced periodicity for the specific case of a/b = 4.45/5.80 of Fig.2a is provided below.

Understanding the direction of motion and reduced periodicity in Fig.2a through the stick-slip displacement.

An interesting aspect that we observed in Fig.2**b** (and Movie 4) is that, during a stick-slip motion, the cluster advances in such a way that one neighbour of a current blue particle becomes the new blue particle after a slip motion. This process continues as long as directional locking is observed. Fig.S8 shows the six shortest displacements $\Delta\mathbf{r}_i$ (i=1,2,3,4,5,6) that can lead to trapping a neighbor particle without rotating the whole cluster. Clearly, the direction of $\Delta\mathbf{r}_4$ has the smallest deviation from that of the external driving force **F**. Therefore, a simple description of the directional locking is that, the cluster takes $\Delta\mathbf{r}_4$ as the slip direction at every stick-slip motion. Consequently, the direction of motion θ_d equals $O(\Delta\mathbf{r}_4)$, the direction of $\Delta\mathbf{r}_4$. Therefore $\theta_d = O(\Delta\mathbf{r}_4) = 60° + O(\Delta\mathbf{r}_3) = 60° + O(\mathbf{b}_1 - \mathbf{a}_1) \approx 60° + O(\mathbf{a}_2/2) = O(\mathbf{a}_1) = \theta_o = 19.1°$. Since $\theta_s = 0°$ here, this agrees with the above expression $\theta_d = \theta_o - \theta_s$. The reduced periodicity is then $|\Delta\mathbf{r}_4| = |\mathbf{b}_1 - \mathbf{a}_1| \approx |\mathbf{a}_2/2| = 2\pi/(\sqrt{3}/2|\boldsymbol{Q}_s|)$ for the specific case of $\boldsymbol{Q}_s = -\boldsymbol{\beta}_1 + 2\boldsymbol{\beta}_2 = -2\boldsymbol{\alpha}_2$. Here the '≈' indicates that we have used the approximation $2\mathbf{b}_1 \approx 2\mathbf{a}_1 + \mathbf{a}_2$. This geometrical approach could also generalize to arbitrary a/b by considering the minimum particle-hole displacement of all the particles within the unit cell of a superstructure.

Possible influence of hydrodynamic interparticle interactions.

Hydrodynamic interactions (HI) can add torques to the clusters which leads to realignment. This effect, if strong enough, could rotate the cluster out of registry and destroy directional locking. The fact that our experimental results are in good agreement with simulations where HI is neglected, suggests that HI play a minor role. The reason for that is twofold. First, typical cluster velocities are quite small. This is seen in Fig.S9, reporting the x-component of the velocities of the three clusters of Fig.4a as a function of the driving force. The measured velocity is below 0.8 μm/s even at the largest driving force. Second, the patterned substrate provides stick boundary conditions for the fluid flow that forces the velocity field of the fluid to vanish at the walls. Since typical particle distances from the lower wall amount only to few nanometers, this leads to a strong screening of HI.