Electronic Supplementary Material (ESI) for Soft Matter.

Seedless assembly of colloidal crystals by inverted micro-fluidic pumping

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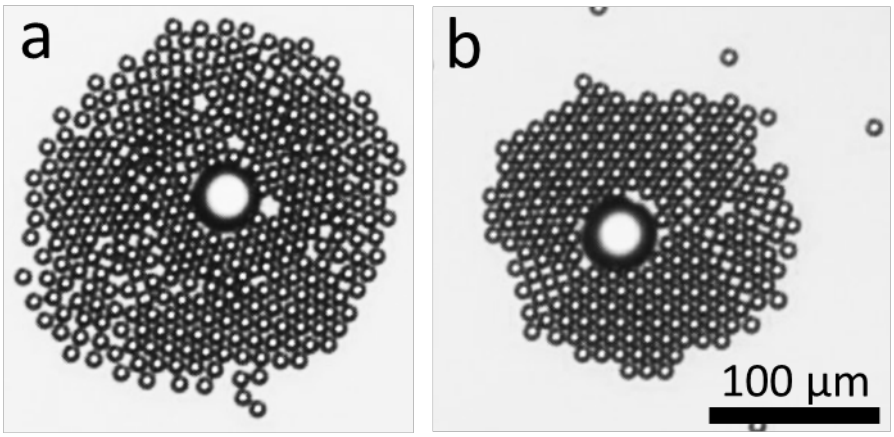

Fig. S1 Low quality colloidal crystal of PS10 in eo-pumping of IEX45: no crystalline order (a) and multi-domain crystal on uncoated substrate due to the incommensurate of the seed and the formed crystal (b). The scale bar in (b) applies for both images.

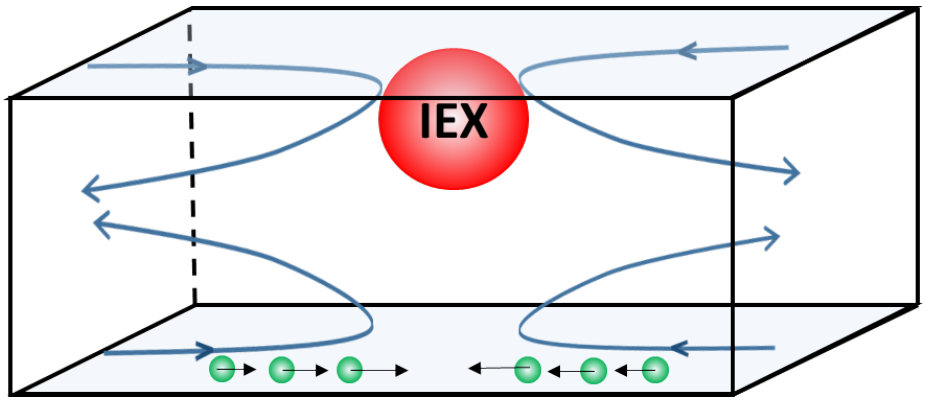

Fig. S2 A schematic drawing of the IEX-based inverted microfluidic platform.

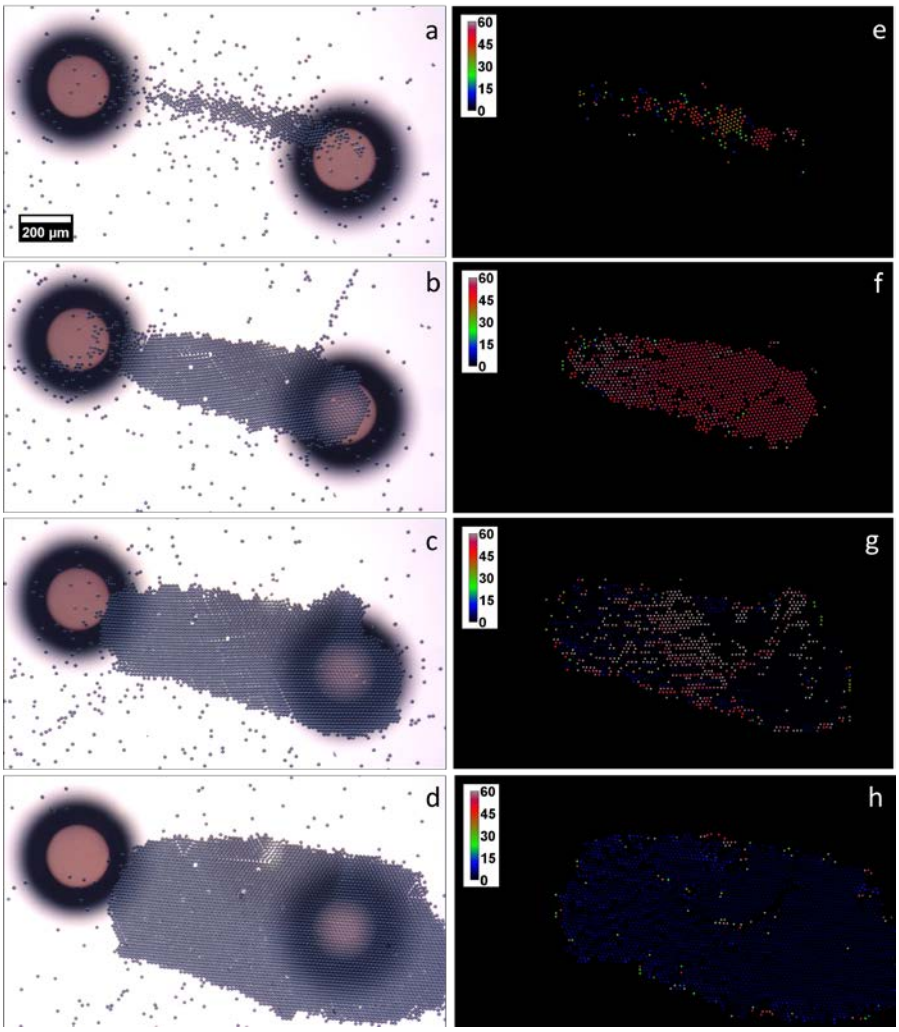

Fig. S3 Typical stages of oriented crystal formation: PS15 in the inverted pump formed by two spaced IEX500 with center-to-center distance of 1116 µm and orientation of 16.9° relative to the horizontal axis. 200 µm scale bar shown in a) applies to all images. (a) At t = 52.5 min, crystal forms between the two IEX500. (b-d) Crystal grows longer along the central line connecting the two IEX; meantime, the crystal grows wider in the direction perpendicular to the central line. In the process, dislocations and grain boundaries develop and anneal. Images were taken at t =122.5 min, 167.5 min and 242.5 min, respectively. The corresponding maps of crystalline particles with $\phi_6 > 0.8$ and the color coded local crystalline orientation relative to the horizontal axis (e-h).

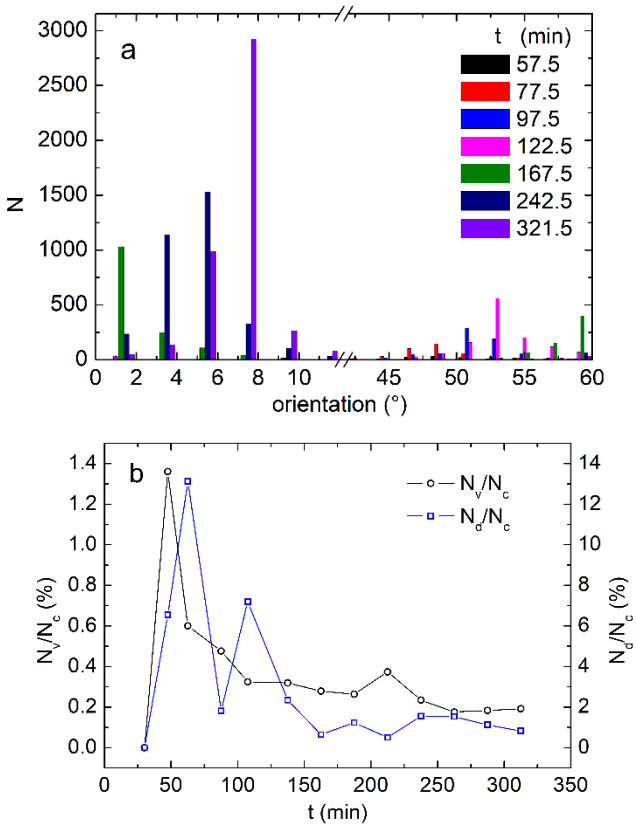

Fig. S4 (a) Histogram of number of crystalline particles with certain orientations relative to the horizontal axis for the images shown in Fig. S3. (b) Fraction of voids N_v/N_c and fraction of particles in dislocations and grain boundaries N_d/N_c as a function of time t from the start of the experiment.

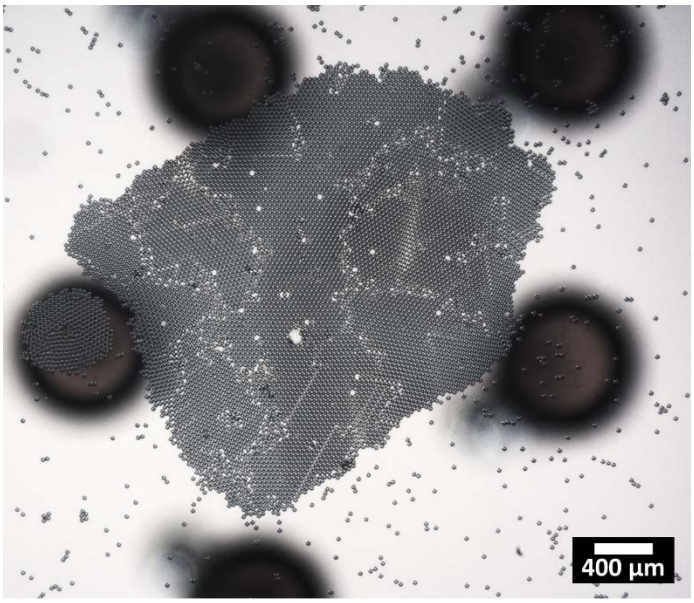

Fig. S5 Patterning pentagon shaped monolayer colloidal crystal using IEX500-based inverted pump from PS15 at *H*= 1 mm. Scale bar as indicated.

Videos:

Video 1. Single IEX660-based inverted pump at *H*= 1 mm. PS7 tracer particles are dragged towards the projected center of IEX660. 14x real time speed. Scale bar as indicated.

Video 2. Single crystal formed in single IEX500-based inverted pump from PS10 at *H*= 1 mm. 600x real time speed. Image size 1217.0x971.6 μm^2.

Video 3. Oriented single crystal formed between two spaced IEX500 (center-to-center distance of 1487 µm, aligned along 30° relative to the horizontal axis)-based inverted pump from PS15 at *H*= 1 mm. 300x real time speed. Image size 1790.2x1342.6 μm^2.

Video 4. Oriented single crystal formed between two spaced IEX500 (center-to-center distance of 1116 µm, aligned along 16.9° relative to the horizontal axis)-based inverted pump from PS15 at *H*= 1 mm. 300x real time speed. Image size 1790.2x1342.6 μm^2.

Video 5. Assembly of PS15 and PS10 in the single IEX-based inverted pump. The assembled colloids are mixed without obvious sorting of the size. 200x real time speed. Image size 2727.3x2045.5 μm^2.