

Electronic Supplementary Information for:

# Highly Parallel Acoustic Assembly of Microparticles into Well-Ordered Colloidal Crystallites

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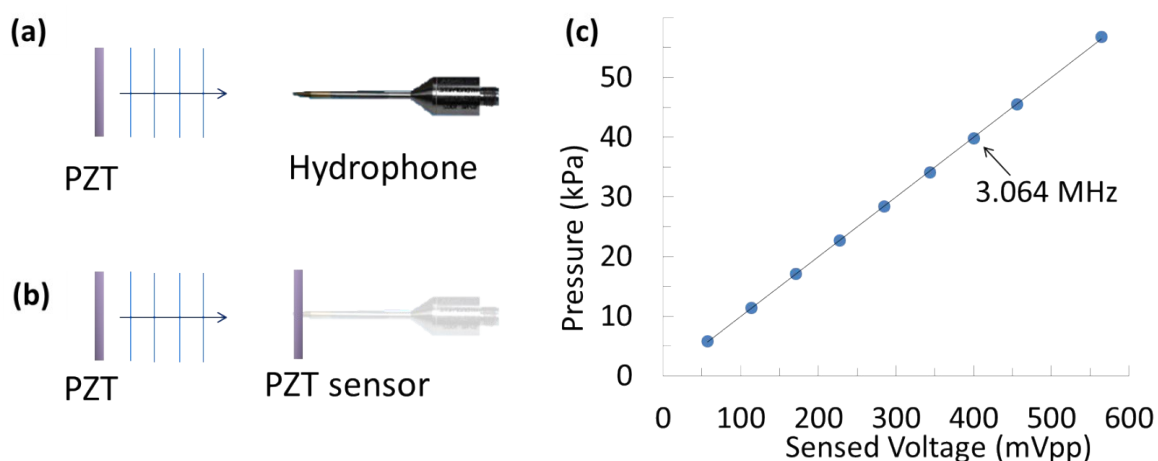
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### 5.1 Pressure Amplitude Determination

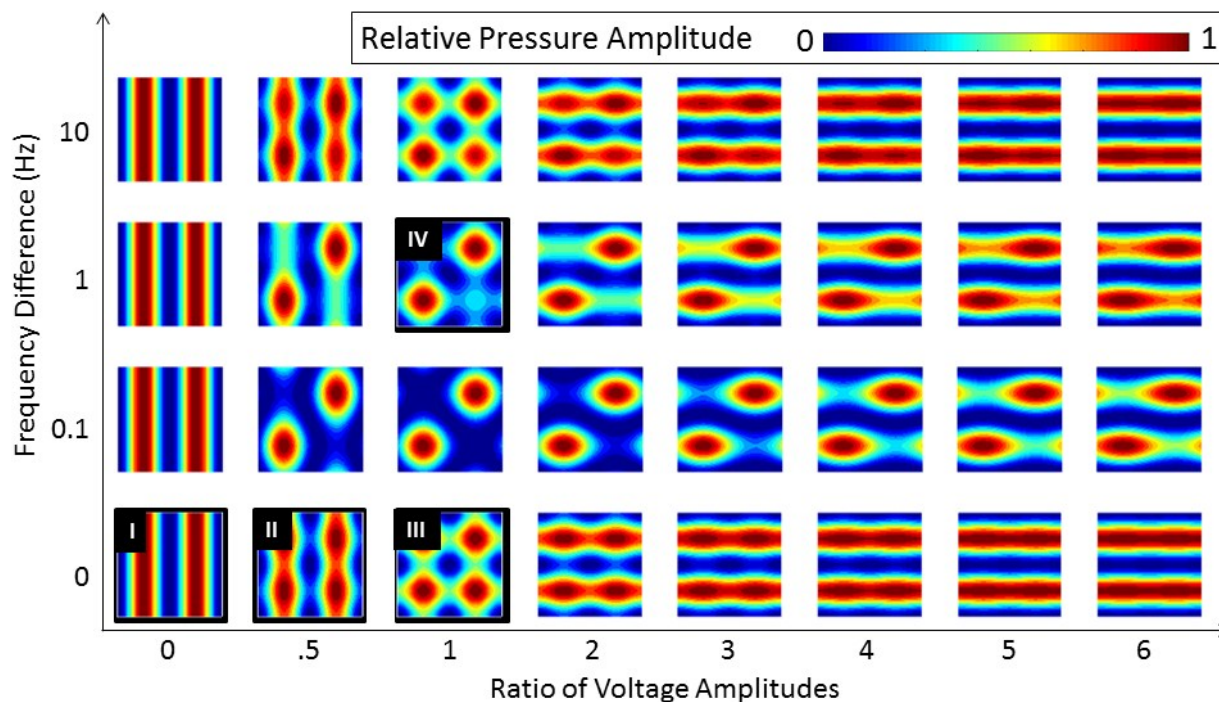
We submerged a driving PZT transducer (30x10x1 mm, 841-WFB, APC International, Ltd.), tethered by two electrical connections encased in epoxy, in a large water bath. We supplied the transducer with AC voltage inputs of 1-10 V peak-to-peak at a constant frequency of 3.064 MHz and measured the pressure amplitudes of the outgoing pressure waves with a hydrophone (ONDA HNA-0400) directly facing the transducer, separated by a distance of 25 mm (Fig. S1(a)). We then used a sensing PZT transducer to measure the corresponding output voltage produced by the first (driving) PZT transducer (Fig. S1(b)) to generate a calibration curve between pressure and PZT voltage (Fig. S1(c)). Using this information, we estimated the pressure amplitudes within the acoustic chamber by placing a sensing PZT transducer along the inside edge of the device (see manuscript, Fig. 2(b)).



**Figure S1.** Method for determining pressure amplitude in the acoustic chamber device. (a, b) A schematic of the experimental setup. (c) Calibration curve of the PZT transducer submerged in water, driven at 3.064 MHz, at a depth of  $\approx 3$  mm and separation distance of 25 mm. Error bars indicating standard error of the mean ( $n = 50$ ) are smaller than graphed points.  $R^2 > 0.99$ .

## 5.2 Variety of Potential Schemes

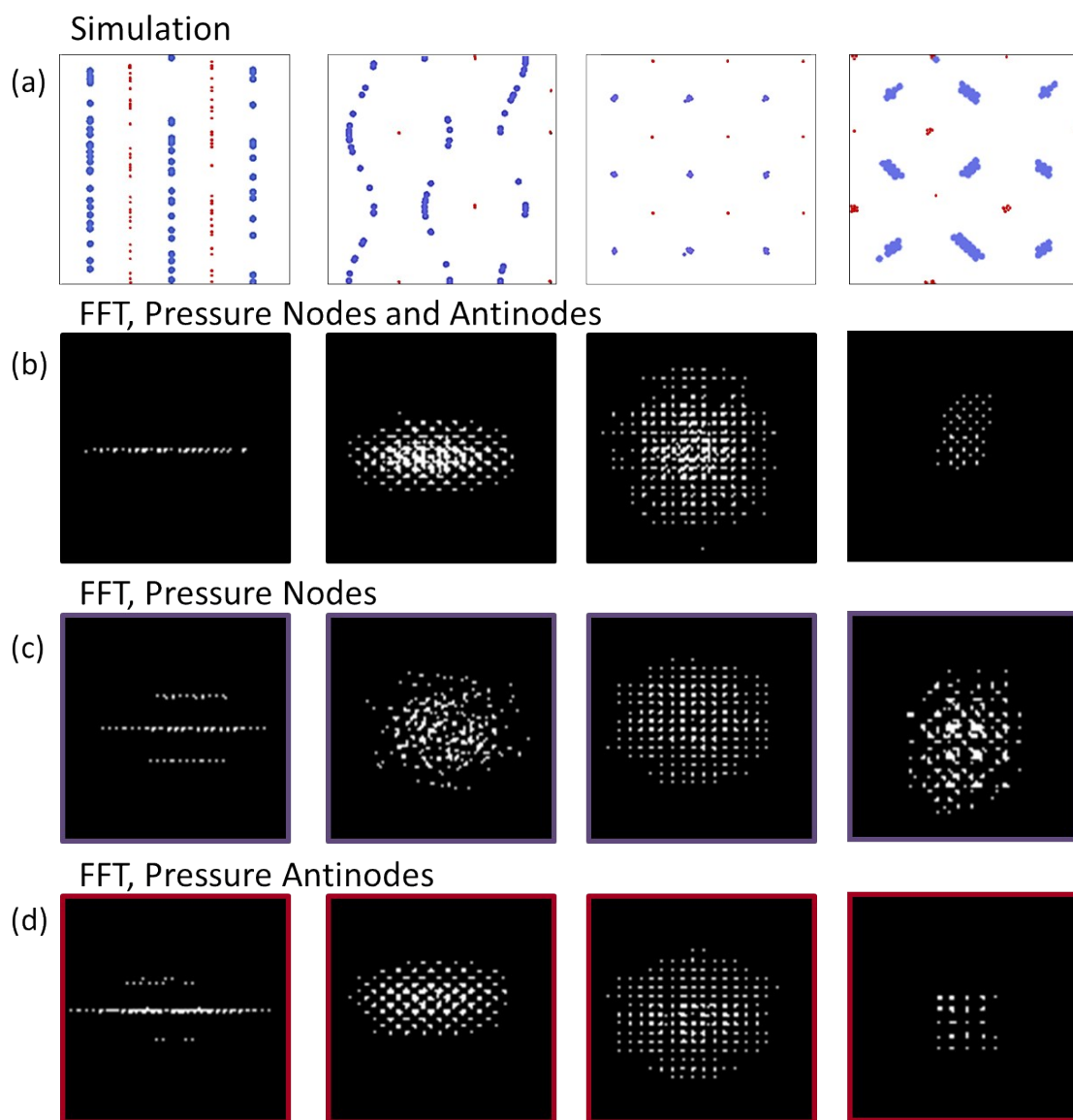
We calculated a variety of conceivable relative pressure amplitudes by altering the frequency and amplitude of the sinusoidal voltage signal applied to the two PZT transducers (Fig. S2). As indicated by the black boxes in Figure S2 below, we selected four schemes for further investigation (see Fig. 4 and Section 3.2 in the text).



**Figure S2.** Schema of time-averaged relative pressure amplitude from acoustic standing waves generated by the two PZT transducers driven at different applied frequencies and sinusoidal voltages. The dark blue regions indicate the pressure nodes and the red regions indicate the pressure antinodes. Black outlines highlight the four schemes selected for further investigation in the manuscript, discussed in Section 3.2 and shown in Figure 4. The voltage ratio is defined as the ratio of the voltage supplied to the  $x$ -transducer to that supplied to the  $y$ -transducer, and the frequency difference is very small compared to the total driving frequency of  $\approx 3$  MHz, so that equal spacing between nodes in the  $x$ - and  $y$ -directions is maintained.

### 5.3 Fast Fourier Transforms of Concentrated Particles

We analyzed the simulation results of the representative mesoscale assembly patterns formed from particles with positive and negative acoustic contrast factors (Fig. S3(a)) using fast Fourier transforms (FFTs) of structures formed along the pressure nodes (Fig. S3(b)), pressure antinodes (Fig. S3(c)) and both the pressure nodes and antinodes of the acoustic standing waves within the device (Fig. S3(d)).



**Figure S3.** Fast Fourier transforms (FFTs) of images of assembled particles from the simulation.

(a) Simulation results (also shown in Fig. 4(e-h) of the manuscript) of particles with positive and

negative acoustic contrast factors collecting along the pressure nodes and antinodes of the standing wave (blue and red particles have positive and negative acoustic contrast, respectively). FFTs of particles assembled along (b) the pressure nodes, (c) the pressure antinodes and (d) both the pressure nodes and antinodes of the acoustic standing waves within the chamber device. The conditions for each column correspond to the same conditions of the columns shown in Figure 4 of the manuscript.

#### 5.4 Stacking Transition

The approximate transition between monolayer and multilayer structures is estimated by a simple energy model, which compares the acoustic potential energy (Eqn. 1) in a particle at a given distance from the node center and the gravitational potential energy necessary to raise it out-of-plane. We predict that stacking (or buckling) occurs when the acoustic potential exceeds the gravitational potential (see Fig. 7(a) in the manuscript for this theoretical relationship plotted as a function of initial particle concentration and pressure amplitude). The theoretical line used in Figure 7(a) is shown below (see Table 1 for a list of variables):

$$\frac{p_0^2 V}{8} \left( 2 \left( 1 - \frac{\rho_m c_m^2}{\rho_p c_p^2} \right) \left( (\cos(kx \cdot x))^2 - 1 \right) - \frac{3 \left( \frac{2\rho_p}{\rho_m} - 2 \right)}{\frac{2\rho_p}{\rho_m} + 1} \sin(kx \cdot x)^2 \cos(\theta)^2 \right) - 2V\rho_p a g = 0$$

#### 5.5 Videos of Particle Acoustophoresis, Assembly and Out-of-Plane Buckling

*Video 1:* Simulation of particles with positive acoustic contrast factors assembling into multiple nodes ( $p_0 = 80$  kPa). A Brownian dynamics simulation developed in MATLAB simulates the assembly of polystyrene particles (10  $\mu\text{m}$  diameter) with a positive acoustic contrast factor (i.e.,

$\phi \approx +0.46$ ) into a 3 x 3 grid of pressure nodes, resulting in the formation of close packed structures. The acoustic standing waves were activated at the beginning of the video and maintained throughout the video. The time of the simulation is indicated at the top of the video. The video repeats three times.

*Video 2:* Simulation of particles with positive acoustic contrast factors assembling into a single pressure node. Polystyrene particles (10  $\mu\text{m}$  diameter) with a positive acoustic contrast factor (i.e.,  $\phi \approx +0.46$ ) assemble into a close packed structure ( $P_o = 100$  kPa) and one particle is forced out-of-plane due to the imposition of higher acoustic pressure amplitudes and concentration compared to the simulation shown in Video 1. Lines in the bottom x-y plane represent the history of the position of the particles. The time of the simulation is indicated in the top, left hand corner of the video. The video repeats three times.

*Video 3:* Experimental separation of particles with positive and negative acoustic contrast factors. A two-dimensional standing wave driven by two orthogonal PZT transducers is activated at the beginning of the video (70 kPa for each PZT transducer). Initially, a solution positive acoustic contrast particles (fluorescent 10  $\mu\text{m}$  diameter cubic SU-8 particles fabricated from photolithography;  $\phi \approx +1.17$ ) and negative acoustic contrast particles (1.5  $\mu\text{m}$  diameter silicone spheres synthesized from a 96:1 molar ratio of dimethoxymethylsilane:tetramethoxysilane using a method described previously;  $\phi \approx -0.88$ )<sup>1</sup> are randomly distributed on the floor of the device. Upon imposition of the acoustic standing waves, the SU-8 cubes and silicone spheres quickly separate to the pressure nodes and antinodes, respectively, to form an arrangement described by Scheme III (see Fig. 4 in the manuscript). The time of the experiment is indicated in the top, left hand corner of the video. The video repeats three times.

*Video 4:* Experimental acoustophoresis of polystyrene particles with one PZT transducer active (Scheme I) and two PZT transducers active (Scheme III). Initially, an acoustic standing wave is applied to a random distribution of particles ( $P_o \approx 100$  kPa). Once the standing wave is formed, polystyrene beads (10  $\mu\text{m}$  diameter) with a positive acoustic contrast factor ( $\phi \approx +0.46$ ) migrate to the pressure nodes to form lines. Once these are formed, a second, orthogonal PZT transducer is activated, creating a two-dimensional standing wave, and particles migrate to the pressure nodes to form a square lattice arrangement. The time of the experiment is indicated in the top, left hand corner of the video.

## References

1. C. W. Shields IV, D. Sun, K. A. Johnson, K. A. Duval, A. V. Rodriguez, L. Gao, P. A. Dayton and G. P. Lopez, *Angew Chem Int Ed Engl*, 2014, **53**, 8070-8073.