

## **SUPPORTING ONLINE MATERIAL**

### **Templated Self-Assembly in Three Dimensions Using Magnetic Levitation**

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## **S.1 Details of the Mag Lev Device**

Permanent NdFeB magnets (Grade N50, shaped as rectangular prisms with dimensions of 2 in  $\times$  2 in  $\times$  1 in, supplied by K&J Magnetics) with a surface field of  $\sim 0.4$  T generated the magnetic field. The magnets were held at a distance of 20-70 mm (the distance between the magnets could be adjusted) in an anti-Helmholtz arrangement (Figure 1a).

## **S.2 Computation**

We used COMSOL Multiphysics (provided by COMSOL AB) to model and visualize the magnetic field and the gradient generated in our device.

## **S.3 Mechanical Agitation**

The motor (RadioShack 3VDC micro vibration motor, item # 273-0107) we used rotates an unbalanced shaft at frequencies up to 16000 rpm with the application of a 3V DC current. It was affixed to the container using epoxy glue; the container was loosely held in the MagLev device so as to not dampen the vibrations.

## **S.4 Permanently joining SA levitating structures**

An emulsion containing photocurable adhesive was prepared by vortex mixing 1:100 v/v adhesive (Norland Optical Adhesive 72):MnCl<sub>2</sub> solution. After the spheres SA, the emulsion was slowly added by hand via a syringe to the container as to not disturb the levitating cluster. We gently mixed the solution by withdrawing and returning liquid from the container. We left the container in the Maglev device undisturbed for five minutes to allow the glue to coat the cluster, and then exposed the solution to UV light for four minutes in a Uvitron Intelli-ray 600W shuttered UV chamber at a lamp distance of 10 cm to crosslink the glue. The objects were then removed and rinsed in water.

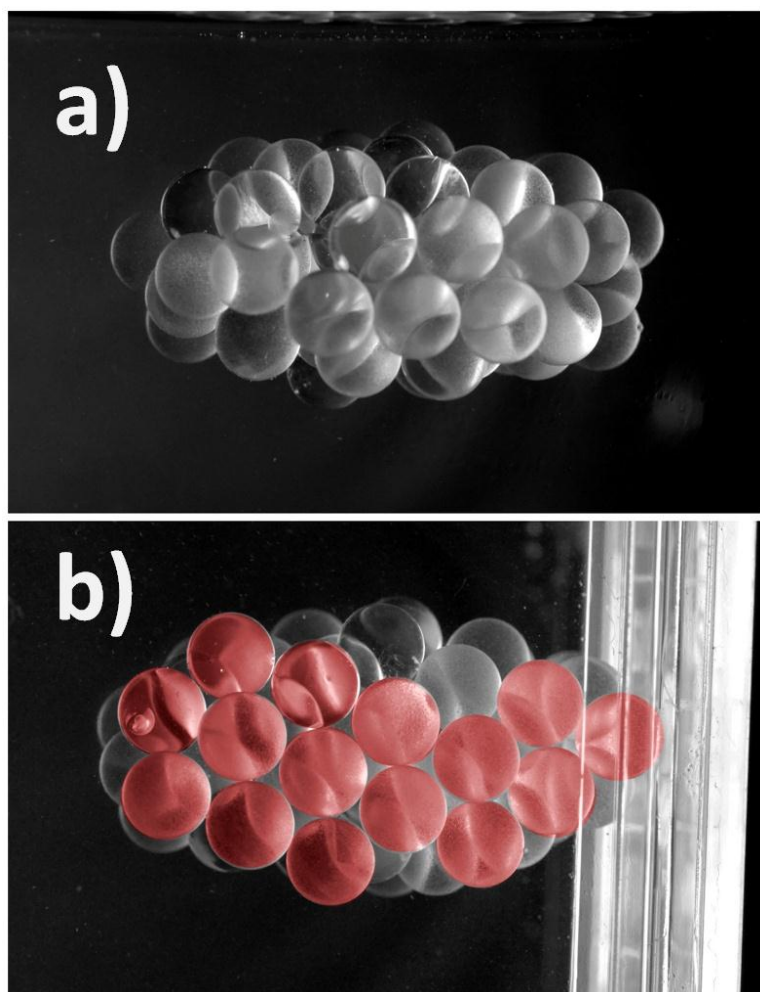
## **S.5 Crystal lattices**

Containers sized to enable the packing of multiple layers in 3D can be used to create 3D crystal lattices (Figure 2, bottom row). The ratio of the size of the levitating components to the dimensions of the container determines the assembly and packing that can be achieved for different containers in 3D (Figure S 2 demonstrates SC, FCC, BCC and monoclinic packing of spheres in containers). Many studies have examined packing of spheres, both theoretical and experimental<sup>32-35</sup>. Packing of levitating spheres in containers largely followed the rules outlined by Wu et al.<sup>34</sup> with respect to the ratio of the diameter of the sphere to the size of the container. The distinguishing feature in our results, however, is that the SA structures are fully suspended in fluid and do not come in contact with the bottom surface of the container.

## **S.6 Preparing complex interlocking objects**

We applied adhesive-backed polytetrafluoroethylene tape (purchased from McMaster Carr) to acrylic sheets and used laser cutting to produce the rectangular slabs with pre-drilled positioning holes for the spheres. The tape is used to control the orientation and levitation height of individual components. The spheres (PMMA, polystyrene and Torlon, purchased from McMaster Carr) with 1/8-in diameter were manually positioned and glued in place using cyanoacrylate glue.

**Figure S 1:** a) A cluster of 60 spheres does not crystallize at any magnet-to-magnet distance, using the magnetic template alone. b) By bringing in the cluster with contact with the wall of the container and applying agitation, we promote fcc ordering, and an observable, crystallized structure emerges. The close-packed  $\{111\}$  plane has been falsely colored red.



**Figure S 2:** Photographs showing different lattices of packed spheres formed by controlling the ratio of the size of the container to the size of the components. a) face-centered-cubic arrangement of  $5/32$  in / 4-mm diameter PMMA spheres in a vertical container, b) simple-cubic arrangement of  $3/16$  in / 4.8-mm diameter PMMA spheres in a vertical container, c) body-centered-cubic arrangement of  $3/16$  in / 4.8-mm diameter spheres with a  $1/16$ -in / 1.6-mm diameter sphere in the center; additional  $1/16$ -in / 1.6-mm diameter spheres tend to cluster around the center region, d) monoclinic arrangement of  $3/16$ -in / 4.8-mm diameter spheres in an angled container. Inner width (wall-to-wall) of the container is 10 mm.

