Supplemental Information

Digital Colloids: Reconfigurable Clusters as High Information Density Elements

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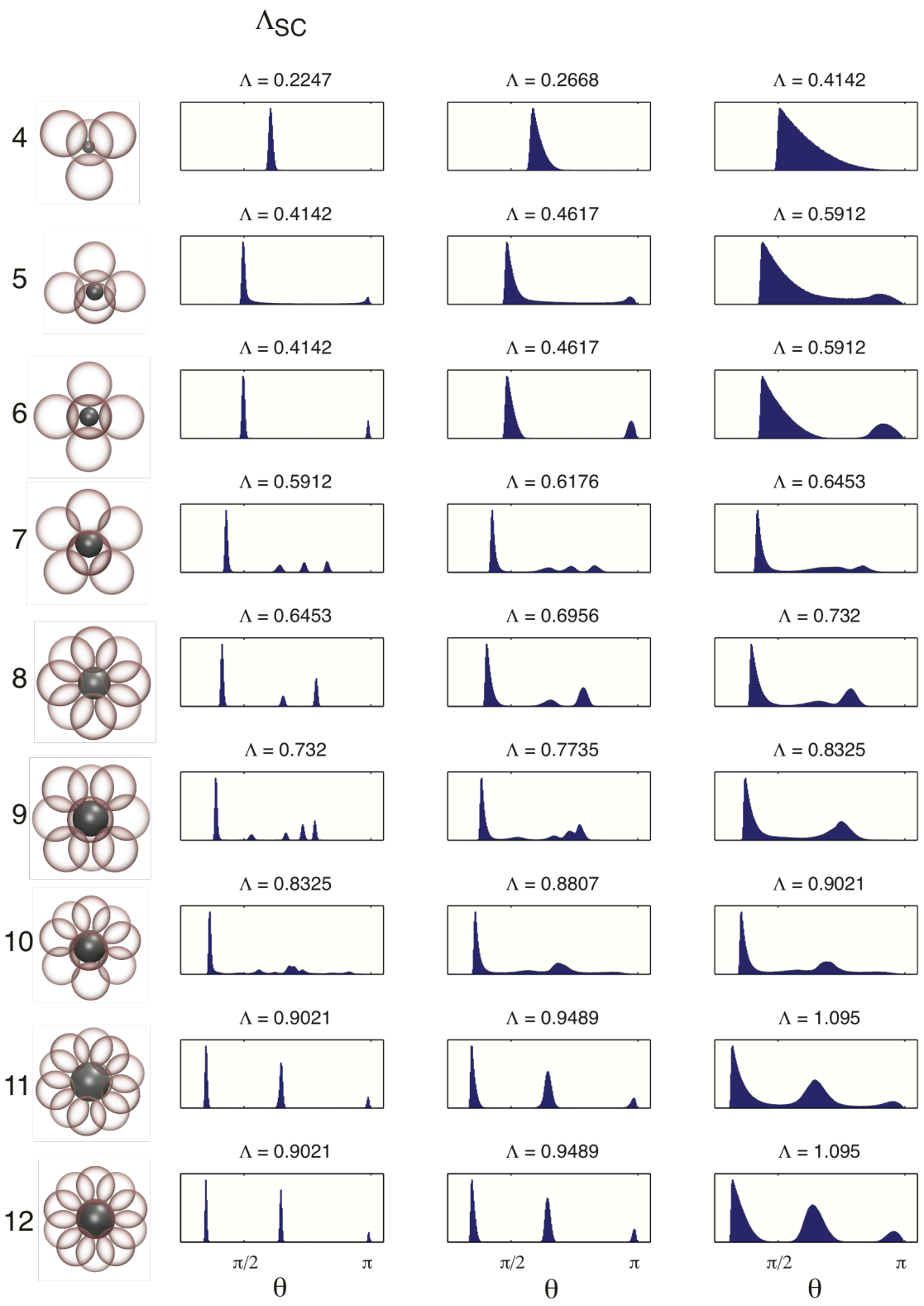

Figure SI-I-1. The distribution of angular displacements n(θ)

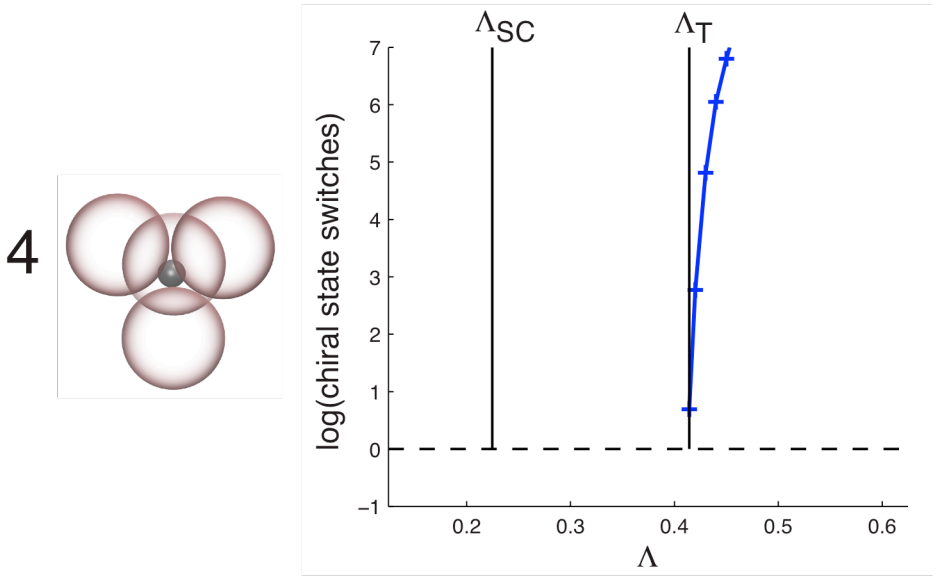

Figure SI-I-2. N = 4. Transitions for N=4 are measured by detecting changes in the chirality of the cluster

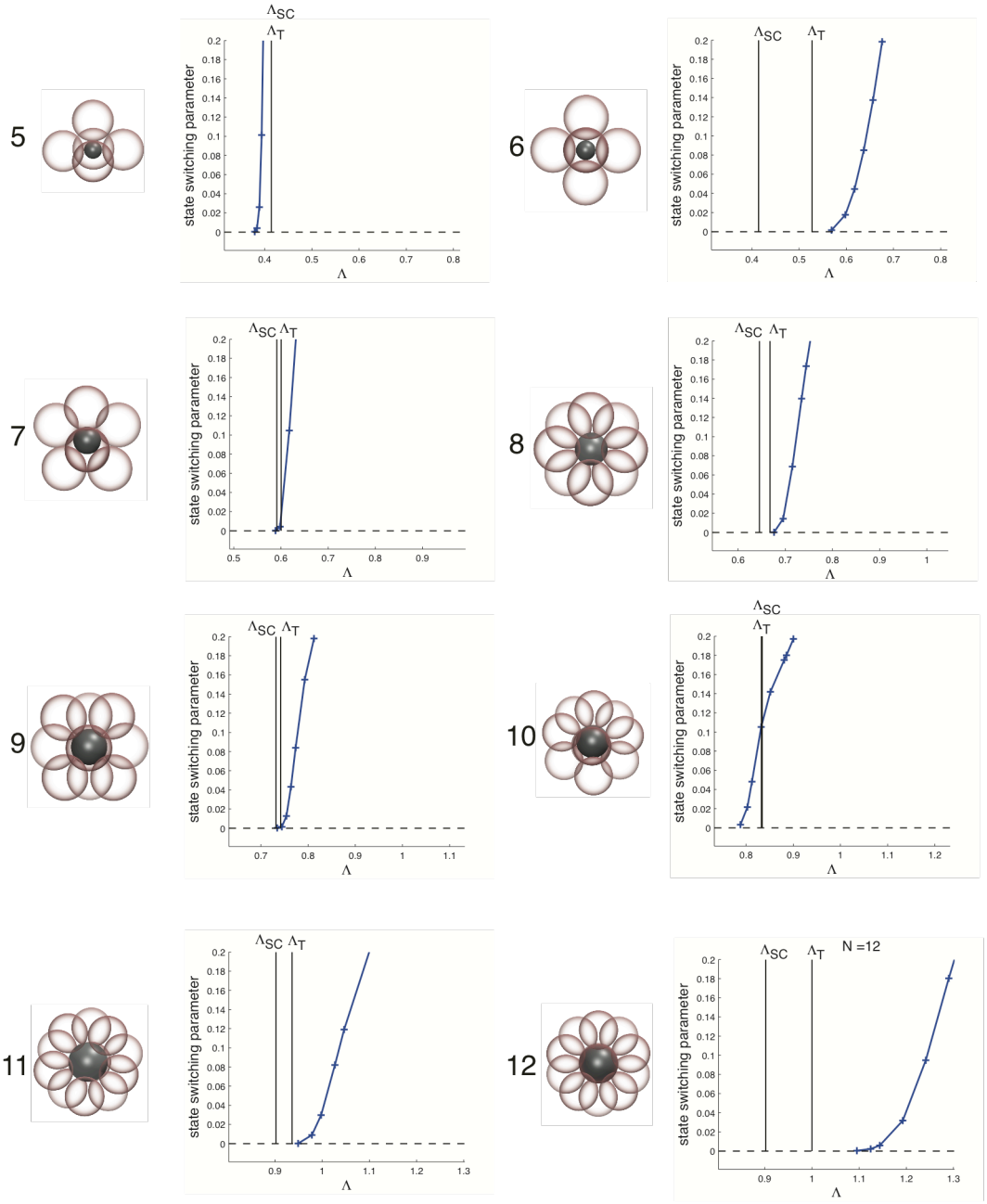

Figure SI-I-3. For N = 5-11 the state switching parameter as a function of Λ. In each figure, as Λ increases, the rate of angular decorrelation also increases.

SI - II - N = 5 cluster has no transition state

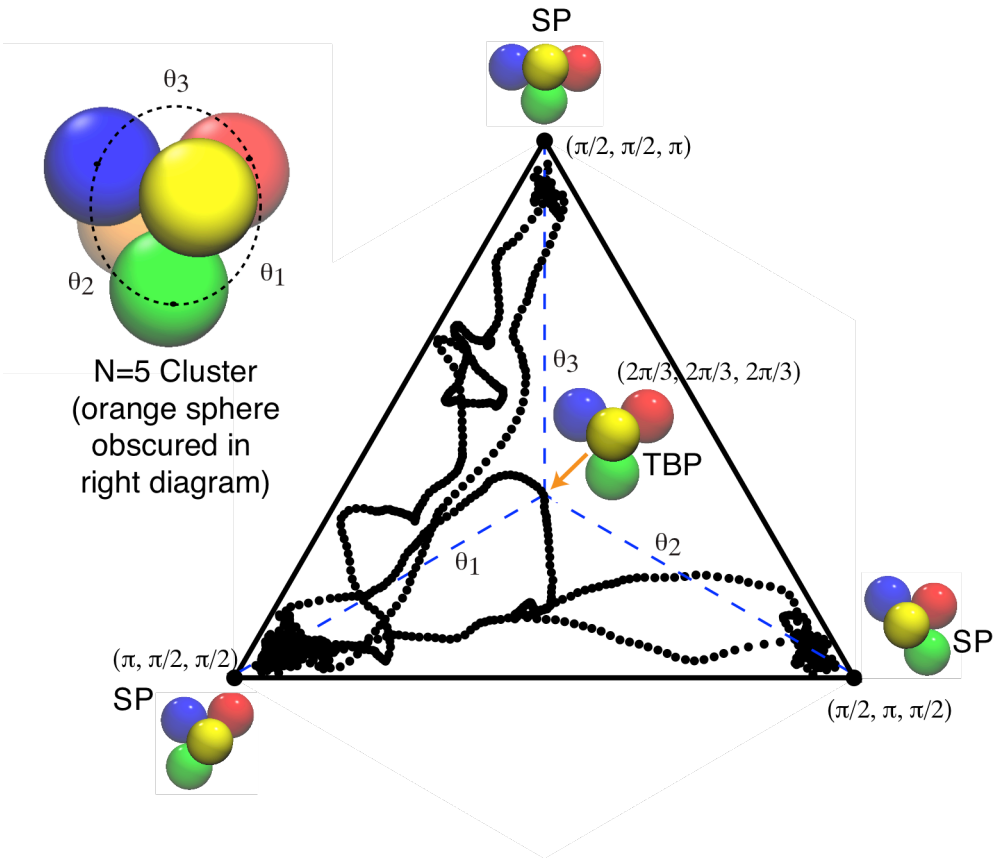

Figure SI-II-1. Four sample transition paths that all started in the bottom left state. The paths pass through no common point.

The N = 5 spherical code solution has a degenerate continuum of structures ranging from a square pyramid (SP) to a triangular bipyramid (TBP) arrangement of spheres. In reference [1], it was shown that entropically, the square pyramid is the preferred structure of the thermodynamic cluster. The square pyramid structure is equivalent to the N = 6 spherical code solution, an octahedron, minus a single outer sphere. If we consider before and after snapshots of an N = 5 cluster going through a transition, we observe that a outer sphere has moved into the available vacancy, and the vacancy is now at the coordinate position the outer sphere de-occupied. However, careful observation of the transition shows that this is not accomplished by the jumping of a single outer sphere. Rather, the cluster enters a metastable set of states, the non-entropically preferred continuum, and then collapses into a nearby square pyramid state.

We demonstrate this in Figure SI-II-1. At its densest, the state of the N=5 cluster can be specified by exactly two numbers, the smallest two angles between outer spheres

on the equator of the cluster. In figure SI-II-1, however, we plot (θ_1, θ_2, θ_3) the three angles between the outer spheres on the equator. This vector is restricted to a planar triangle. When the cluster is in a square pyramid state, the state of the system is near a corner of the triangle. Illustrations of the three SP states are shown. The rest of the triangle represents the phase space of all the states that are not a square pyramid. The triangular bipyramid is the center of the phase space. In Figure SI-II-1, we show the path of the cluster through state space during three transitions. There is no single point that the paths pass through. Rather the cluster enters the metastable middle of state space and then collapses to a nearby corner of state space.

[1] C. L. Phillips, E. Jankowski, M. Marval, and S. C. Glotzer, Phys. Rev. E 86, 041124 (2012).

SI – III HOOMD script for N=12 cluster

```
from hoomd_script import *
import math

N_B =12;

# Ratio
CPoverHP = 1.05

# Temperature
T=0.1;

# Need to Calculate the Appropriate Radial Offset of the
for the ratio provided
# HP diam will grow to 3.0786 in Barker-Henderson units.
The diameter will be 3.0 in LJ units.

Hdiam = 3.0786
Cdiam = CPoverHP*Hdiam
radial_offset = (Cdiam + Hdiam)/2.0;

halo_initial_diameter = 1.0;
halo_diameter = 3.0;
central_diameter = Cdiam + (Hdiam - halo_diameter);

# Distribute the HP spheres on a spiral around CP.
inc =  math.pi  * (3 - math.sqrt(5.));
off = 2. / N_B;

initial_positions = []

for k in range(0,N_B):
    y = k * off - 1 + (off / 2.0);
    r = math.sqrt(1 - y*y);
```

```
    phi = k * inc;
    initial_positions.append((math.cos(phi)*r*radial_offset,
y*radial_offset, math.sin(phi)*r*radial_offset))


##############   INITIATE SIMULATION ###############
system = init.create_empty(N=N_B+1,
box=data.boxdim(L=20),particle_types = ['H','C'])
for k in range(0,N_B):
    system.particles[k].type = 'H'
    system.particles[k].diameter = halo_initial_diameter
    system.particles[k].position = initial_positions[k]

system.particles[N_B].type = 'C'
system.particles[N_B].diameter = central_diameter
system.particles[N_B].position = (0,0,0)

xml = dump.xml()
xml.set_params(all=True)
xml.set_params(image=False)
xml.write(filename='Init.xml')

halo = group.tags(tag_min=0,tag_max=11)

#Pairwise Interaction Force
lj = pair.slj(r_cut = 2**(1.0/6.0),d_max =halo_diameter)
lj.pair_coeff.set('C', 'C', epsilon=0.0, sigma=1.0)
lj.pair_coeff.set('H', 'H', epsilon=0.1/T, sigma=1.0)
#Interaction is independent of Temperature
lj.pair_coeff.set('C', 'H', epsilon=0.0, sigma=1.0)

lj.set_params(mode="shift")

#Constrain Sphere
constrain.sphere(group=halo, P=(0,0,0), r=radial_offset)
integrate.mode_standard(dt=0.001)

bd=integrate.bdnvt(group=halo, T=T, seed=54321)

#Allow to find a random arrangement
run(int(2e5))

# Grow the Halo Spheres
for t in range(0,1001):
    for k in range(0,N_B):
        system.particles[k].diameter = t*(halo_diameter-
halo_initial_diameter)/1000 + halo_initial_diameter
    run(100)
```

```
# Equilibrate
run(int(1e4))

xml.write(filename='Small.xml')

#Save Data
filename = 'trajectory_' + str(CPoverHP)+'.dcd'
datadcd = dump.dcd(filename=filename, period=1e6, overwrite
= True)

run(10e8)
```