Electronic Supplementary Material (ESI) for Soft Matter.


# Supplementary file: Effect of Confinement and Topology: 2-TIPS vs MIPS

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October 3, 2023

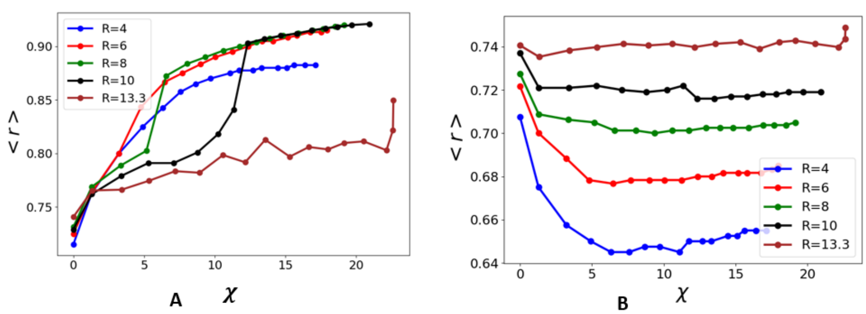


Figure 1: At $\rho^* = 0.8$ A) The plot of mean radial position of the cold particles versus activity $\chi$ for different values of $R$. The mean radial position $< r >$ of cold particles increases with activity $\chi$ for $R = 4\sigma$ to $10\sigma$ indicating that higher number of cold particles move near wall at high activity leading to radial phase separation. For $R = 13.36\sigma$, $< r >$ of cold particles does not reach a maximum value but saturates to a lower value implying that phase separation is not completely radial. B) The plot of mean radial position of the hot particles versus activity $\chi$ for different values of $R$. The mean radial position $< r >$ of hot particles decreases with activity $\chi$ implying that phase separation forces the hot particles to remain in interior of sphere away from wall.

## 1 Mean radial position of hot and cold particles under spherical confinement

We have calculated the mean radial position of all cold particles $< r >$ (normalized it by dividing it by radius R of the confining sphere) as a function of activity $\chi$ for all radii $R$ as shown in fig 1A. If the value of $< r >$ is closer to 1, it indicates that majority of cold particles are near the wall. For small radius of

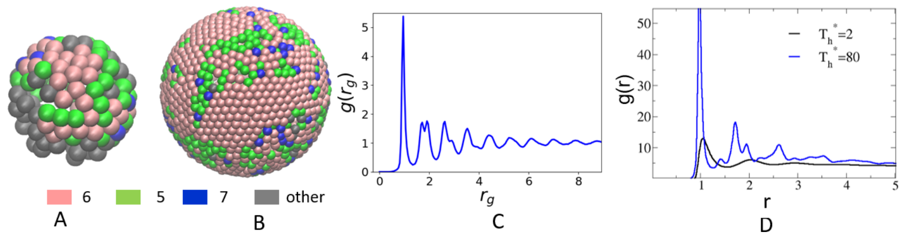


Figure 2: A) and B) Snapshots of cold clusters in the non-equilibrium system inside spheres of radius $R = 4\sigma$ and $10\sigma$ respectively when $\rho^* = 0.8$ and $T_h^* = 80$. The particles are colored based on the number of nearest neighbours. We can clearly see that fraction of particles with six neighbours increases as we move from $R = 4\sigma$ to $10\sigma$. C) The radial distribution function of cold particles inside sphere of radius $R = 10\sigma$ when $\rho^* = 0.8$ and $T_h^* = 80$, calculated using geodesic distance $r_g$ between particles shows crystal structure similar to 2D hexagonal lattice. D) The 3D radial distribution of cold particles inside sphere of radius $R = 13.36\sigma$ when $\rho^* = 0.8$ and $T_h^* = 80$ shows crystalline order similar to bulk phase separation.

| R | 5 nn | 6 nn | 7 nn |
|---|---|---|---|
| 4 | 0.31 | 0.33 | 0.048 |
| 6 | 0.33 | 0.48 | 0.076 |
| 8 | 0.23 | 0.64 | 0.088 |
| 10 | 0.183 | 0.71 | 0.087 |

Table 1: The table contains the fraction of particles in the cold cluster having 5,6 and 7 nearest neighbors (nn), for radius $R = 4\sigma$ to $10\sigma$ when $\rho^* = 0.8$ and $T_h^* = 80$.

R= 4,6 and 8$\sigma$, the mean radial position $< r >$ of cold particles increases continuously with activity $\chi$ and saturates to a value near the wall implying that the majority of cold particles are present near the periphery. For intermediate radius $R = 10\sigma$, the mean radial position $< r >$ of cold particles reaches its maximum value only for high activity. For large radius $R = 13.36\sigma$, the mean radial position $< r >$ of cold particles doesn't reach its maximum value even for high activities. So, smaller the radius of sphere and higher the activity, the higher the affinity of cold particles to be near the wall. Fig 1B shows plot of the mean radial position of all hot particles $< r >$ as a function of activity $\chi$ for all radius $R$. For all radii we observe that the mean radial position $< r >$ of hot particles (Fig 1B) decreases with activity and reaches a constant value as hot particles move into the interior away from the wall.

## 2 Structure of phase-separated cold cluster under spherical confinement.

Mixture of hot and cold LJ particles in bulk at high activity phase separate into crystalline cold clusters and a low-density region containing hot particles in gaseous phase. Cold particles of the phase separated system under spherical confinement also form two kinds of crystalline structures depending on the nature of phase separation: a 2D spherical shell of particles arranged in hexagonal lattice and a 3D cluster of cold particles with HCP and FCC arrangement. For radii in range of $R = 4\sigma$ to $10\sigma$ at density $\rho^* = 0.8$ and $T_h^* = 80$, the 3D system of hot and cold mixture leads to the formation of 2D spherical shell of cold particles near the periphery of the sphere. We investigate the arrangement of cold particles in the shell by calculating the number of nearest neighbors (nn) for each particle. Table 1 gives the fraction of particles in the cold cluster having 5,6 and 7 nearest neighbors (nn), for radius $R = 4\sigma$ to $10\sigma$ when $\rho^* = 0.8$ and $T_h^* = 80$. The number of particles with 6 (5) nearest neighbors increases(decreases) as the radius of the confining sphere is increased. Fig 2A and B shows the snapshots of the cold cluster where the particles are colored based on the number of nearest neighbors when $\rho^* = 0.8$ and $T_h^* = 80$ inside spheres of radius $R = 4\sigma$ and $10\sigma$ respectively. We can clearly see that $R = 10\sigma$ has larger domains of particles with six nearest neighbors compared to $R = 4\sigma$. The particles in cold cluster show structure similar to that of an equilibrium system of particles on the surface of a sphere at high density showing hexagonal lattice with defects. So we calculate radial distribution function (RDF) g($r_g$) of cold particles using geodesic distance $r_g$ between the particles. The RDF g($r_g$) at the geodesic distance $r_g$ from the reference particles is calculated as

$$g(r_g) = \frac{N_{r_g}}{2\pi R^2 sin\theta \Delta\theta \rho} \tag{1}$$

where $N_{r_g}$ is the number of particles in the spherical ring at distance $r_g = R\theta$ with area $2\pi R^2 sin\theta\Delta\theta$. Fig2C shows RDF of cold particles inside sphere $R = 10\sigma$ when $T_h^* = 80$ using geodesic distance $r_g$ between them. The g($r_g$) shows splitting of second peak at $\sqrt{3}\sigma$ and $2\sigma$ resembling RDF in a 2D hexagonal lattice.

Fig2D shows 3D RDF g(r) of cold particles inside sphere $R = 13.36\sigma$ at $\rho^* = 0.8$. The RDF of cold particles show liquid like order when $T_h^* = 2$ and crystalline order when $T_h^* = 80$ similar to bulk phase separation.

## 3 Structure of cold cluster of lj particles on spherical surface

Perfect crystals cannot be formed on the surface of a sphere due to the curvature. The dense region of the non-equilibrium system on the spherical surface also shows the appearance of defects. The particles in the dense region arrange themselves in a hexagonal pattern. Therefore most particles have a coordination number of 6. But due to curvature, the co-ordination number of some point particles are other than 6 (namely 5 and 7). These two types of defects often stay together to form defect lines (fig 3)

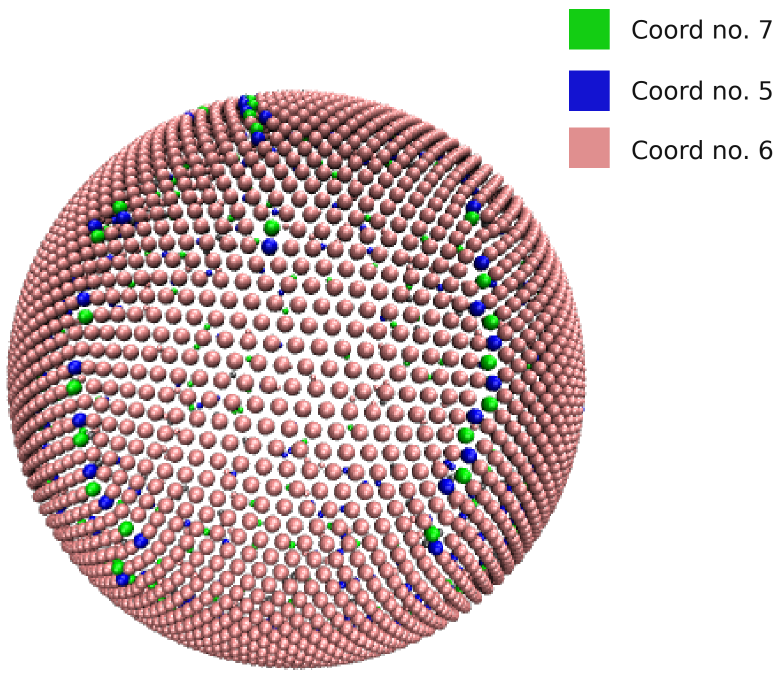


Figure 3: The defect points observed for the cold domain on the surface of the sphere. The points are color coded according to the number of their nearest neighbour, as shown in the figure. We observe that defects with co-ordination number 5 and 7 stay close to each other.

## 4 Additional Figures

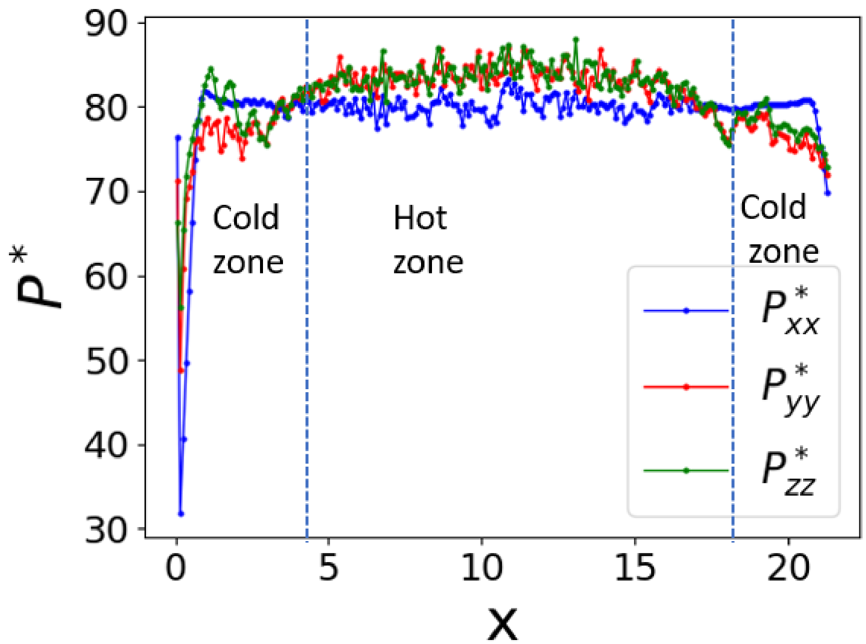


Figure 4: Variation of Normal pressure $P_N$=$P^*_{xx}$ and tangential pressure $P_T$=$P^*_{yy}$=$P^*_{zz}$ along $\hat{x}$ direction perpendicular to interface between hot and cold particles when $\rho^* = 0.8$ and $T^*_h = 80$ in bulk simulation. We can see that in the cold zone, the tangential pressure is lower than normal pressure