# Aggregation Processes in Systems of Fractal Objects

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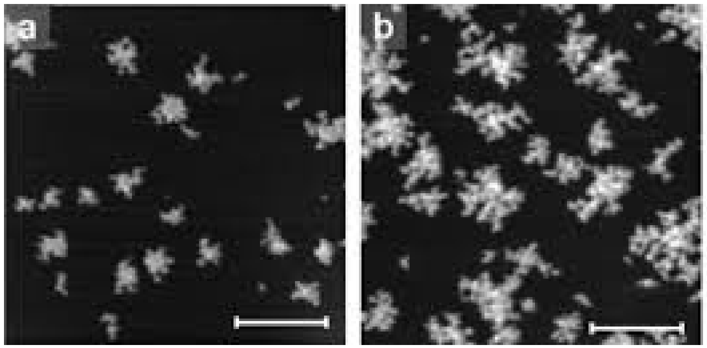
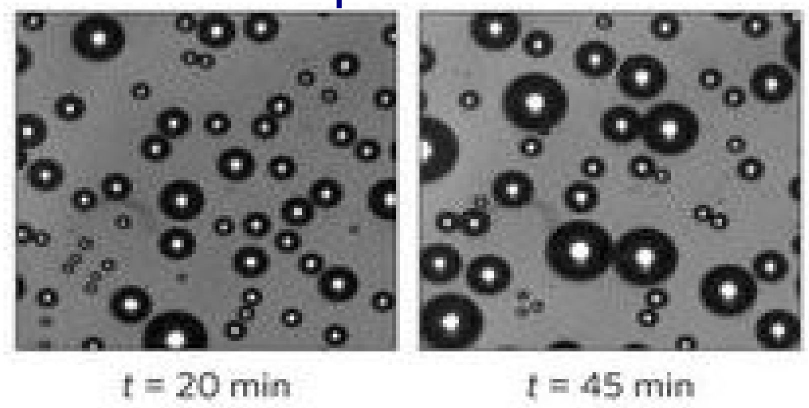
Plan: Introduction
Ballistic Aggregation in Hard Sphere model
in Vapor-Solid Phase Transition
Aggregation in a model Active Matter system
Conclusion.

Ice




Heidom et al. (CPL)

Liquid water

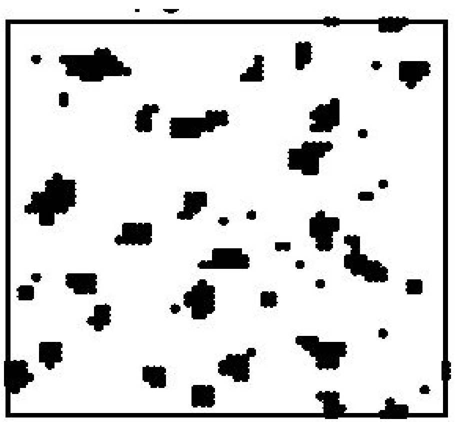



Mouterde et al. (Nature Materials)

## Possible ways of growth in disconnected morphology:

Droplets consist of small particles.

Particles from smaller droplets get detached to be deposited on a bigger droplet.



Droplets **themselves move** and undergo ``sticky Collisions'' with each other to form bigger droplets.

Motion of droplets/clusters can be diffusive, ballistic, … .

**Ballistic aggregation:** Materials Science, Granular Matter, Astrophysics, … .

Carnevale et al., Phys. Rev. Lett. (1990).

Hansen and Trizac, J. Stat. Phys. (1996)

Trizac and Krapivsky, Phys. Rev. Lett. (2003).

J. Blum, Astronomy and Astrophyics (2010).

**2 types:**

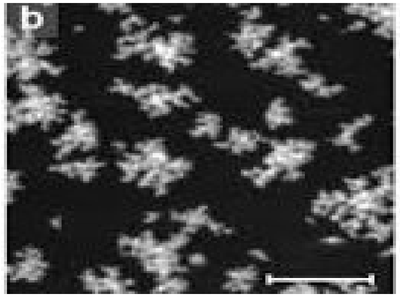
Ballistic deposition – seed is fixed (vapor deposition on a substrate, etc.).


Source: ERCIM

Ballistic aggregation: all clusters move (translation and rotation) and collide.

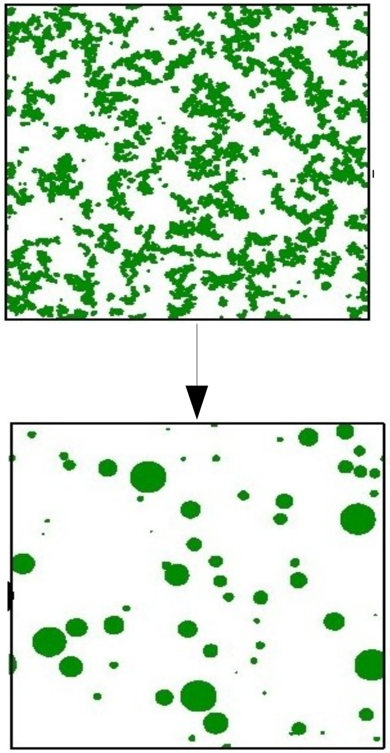

shutterstock.com

**Fractality is usually a natural outcome.**




Heidom et al. (CPL)

Typically simulations are done with ``sticky'' hard spheres – fractal structures are expected.



S. Paul and SKD, Phys. Rev. E (2017)

In event driven simulations, it's necessary to keep track of exact locations as well as orientations of fractal clusters and identify exact points of contact during collision – technically challenging – so spherical cluster approximation becomes necessary.

**Theory:** M. Smoluchowski (1916)
G.F. Carnevale, Y. Pomeau and W.R. Young, PRL (1990)
Hansen and Trizac, J. Stat. Phys. (1996)

$$\frac{dn}{dt} = -Collision-cross-section \times v_{rms} \times n^2$$

$$Collision-cross-section \sim M^{\frac{d-1}{d}} \qquad n \propto 1/M \qquad v_{rms} \sim M^{-1/2}$$

$$\frac{dM}{dt} \sim M^{\frac{d-2}{2\mathrm{d}}} \qquad M \sim t^{\beta}; \ \ \beta = 2\mathrm{d}/(d+2)$$

In event driven simulations … spherical cluster approximation is necessary.

Hansen and Trizac, J. Stat. Phys. (1996).
Trizac and Krapivsky, Phys. Rev. Lett. (2003).
S.N. Pathak, Z. Jabeen, D. Das and R. Rajesh, Phys. Rev. Lett. (2014).
S. Paul and SKD, Phys. Rev. E (2017).

SO the role of fractality in growth is NOT properly known because of spherical structural approximation – for same average mass, collision cross-section should be higher for (rotating) fractal clusters.

Time step driven MD (TDMD) can take care of such difficulty – events occur naturally.

But TDMD applies usually to non-hard sphere kind of systems, like Lennard-Jones inter-particle/atomic potential.

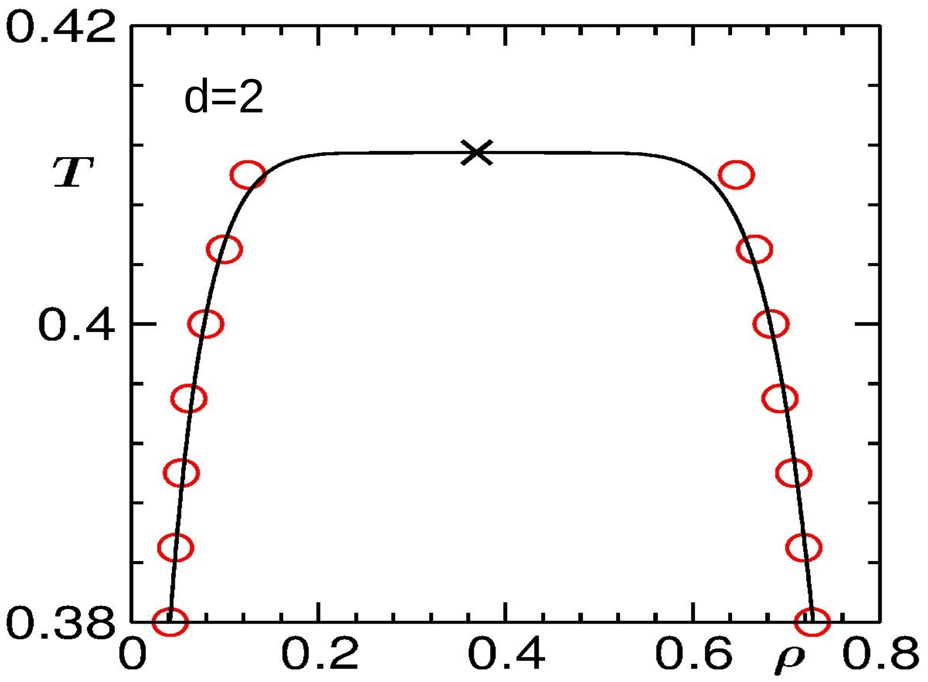
How will such particles stick and keep fractality?

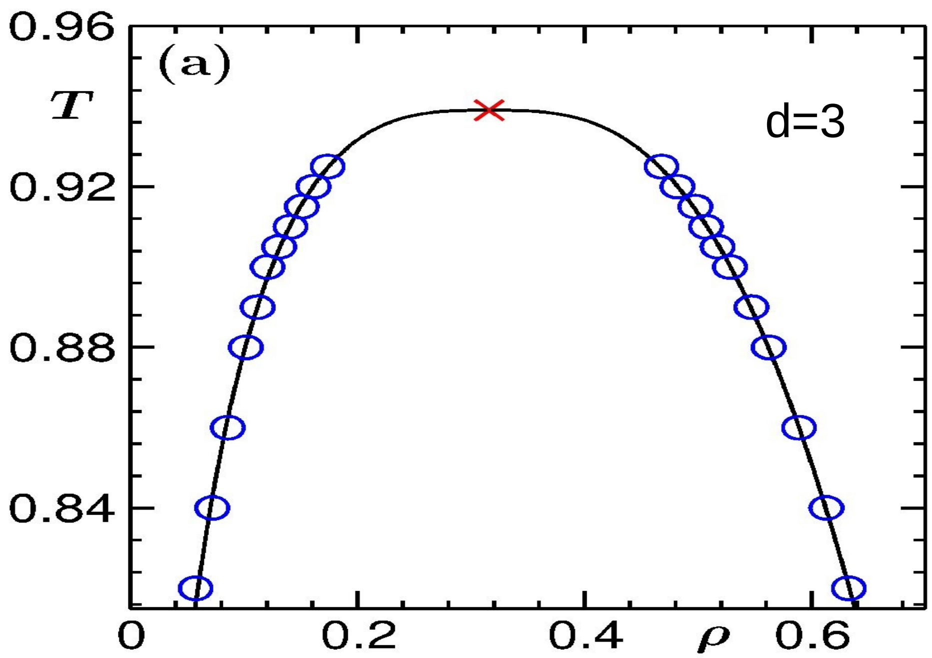
Even if clustering occurs they will try to gain spherical structure to minimize (interfacial) free energy. ??

Typically one uses Model H, Lattice Boltzmann, Molecular Dynamics.

***Our Model:*** $\varphi(r) = 4\epsilon[(\frac{\sigma}{r})^{12} - (\frac{\sigma}{r})^{6}]$ LJ potential

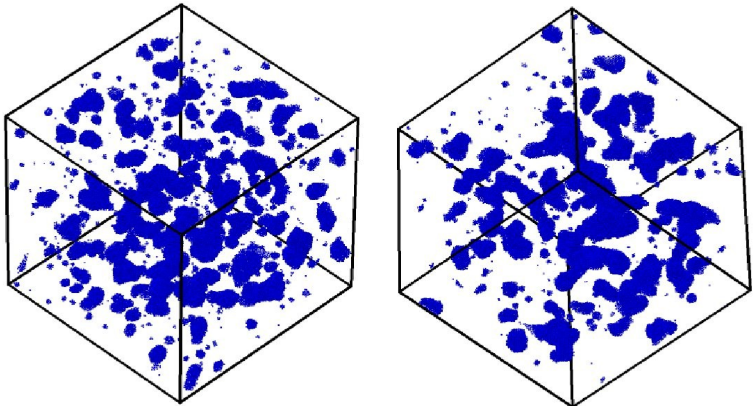
Phase behavior: Monte Carlo simulations.




J. Midya and SKD: J. Chem. Phys. (2017).
J. Chem. Phys. (2017).

Kinetics?: Molecular dynamics simulations with Nose-Hoover thermostat.



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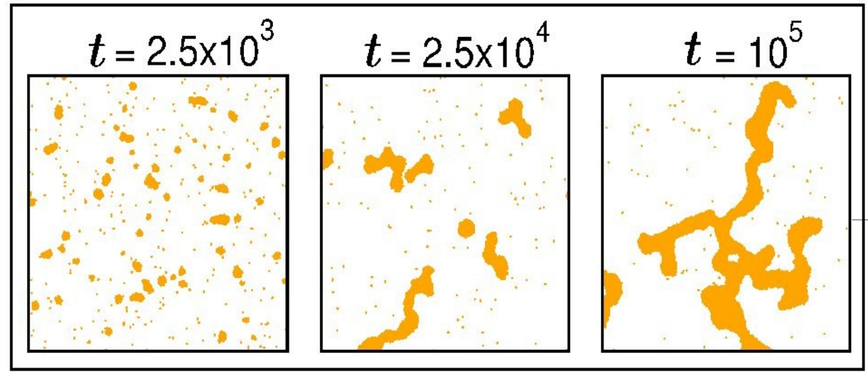
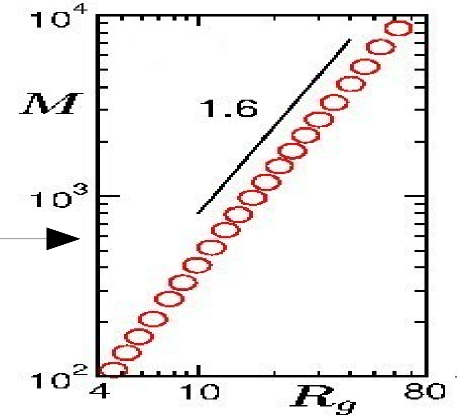
S. Roy



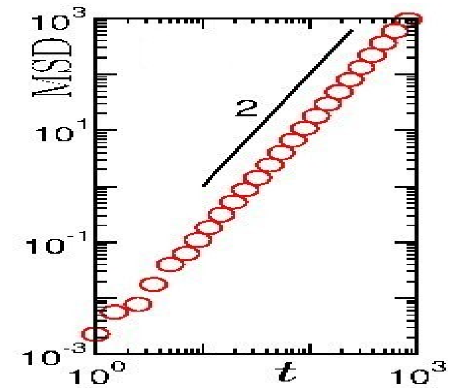

J. Midya



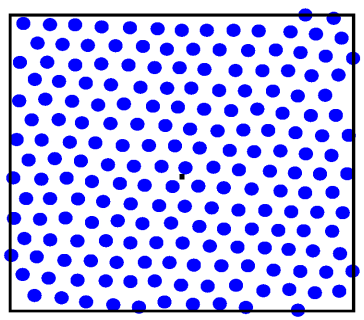

$$M \sim R_g^{d_f}$$
$$d_f = 1.6$$

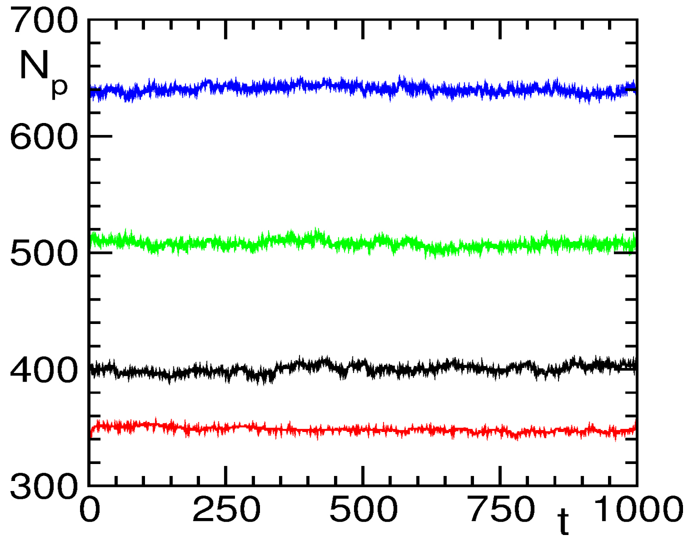
**Fractality**: due to two well separated time scales.

1) (fast) ballistic motion of the clusters (negligible resistance from low density/inviscid vapor background).



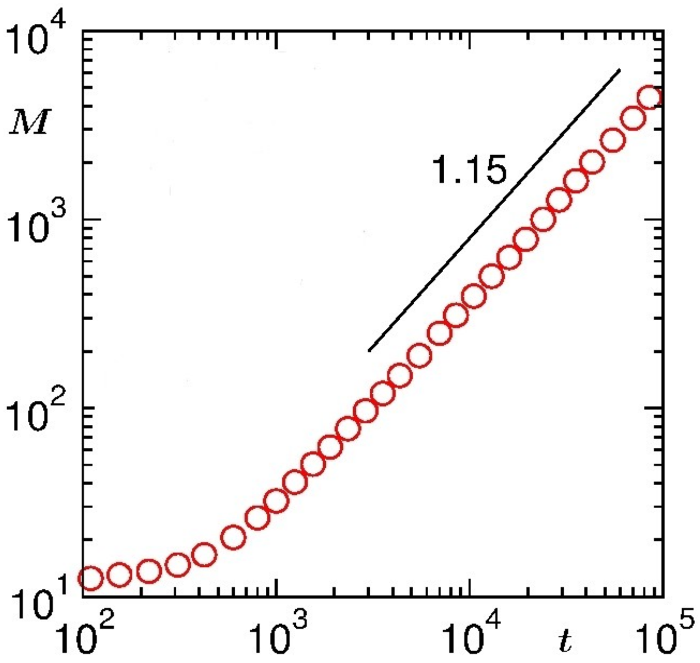

2) (extremely) slow relaxation inside the clusters due to rigidity in the ``solid'' phase.






Particle diffusion mechanism (practically) does not contribute to the growth.

Problem of ballistic aggregation.




But, for ballistic aggregation:

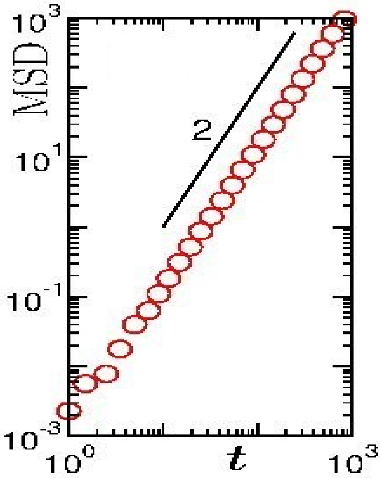
$$M \sim t^{\beta}; \quad \beta = 1.15 \qquad\qquad M \sim t^{\beta}; \ \ \beta = 2d/(d+2)$$

$$d = 2: \ \ \beta = 1$$

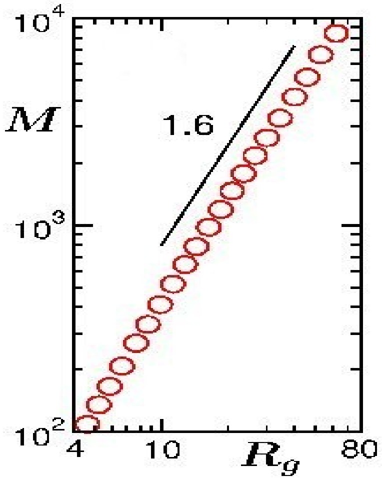
Discrepancy due to fractality?

**<u>YES.</u>**

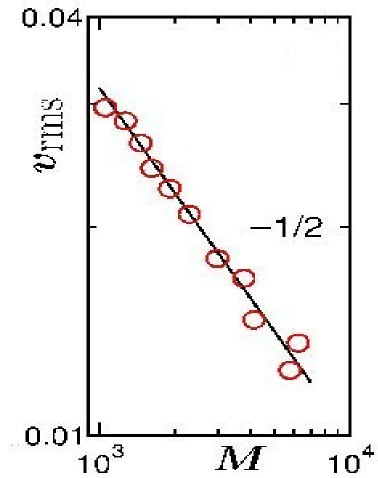
Ballistic motion




Fractal structure




Uncorrelated velocity




**Problem**:
Ballistic Aggregation of Fractal Objects

Theory: $$\frac{dn}{dt} = -Collision\ \ cross\ \ section \times v_{rms} \times n^2$$

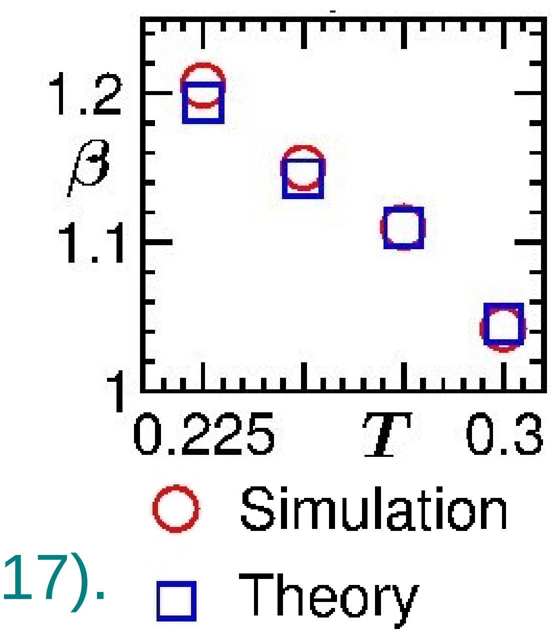
M. Smoluchowski (1916)

$$Collision-cross-section \sim R_g^{d-1} \qquad R_g \sim M^{1/d_f}$$

$$v_{rms} \sim M^{-1/2} \qquad n \sim 1/M$$

Solution: $M \sim t^{\beta};\ \ \beta = 2d_f/(3d_f - 2d + 2)$

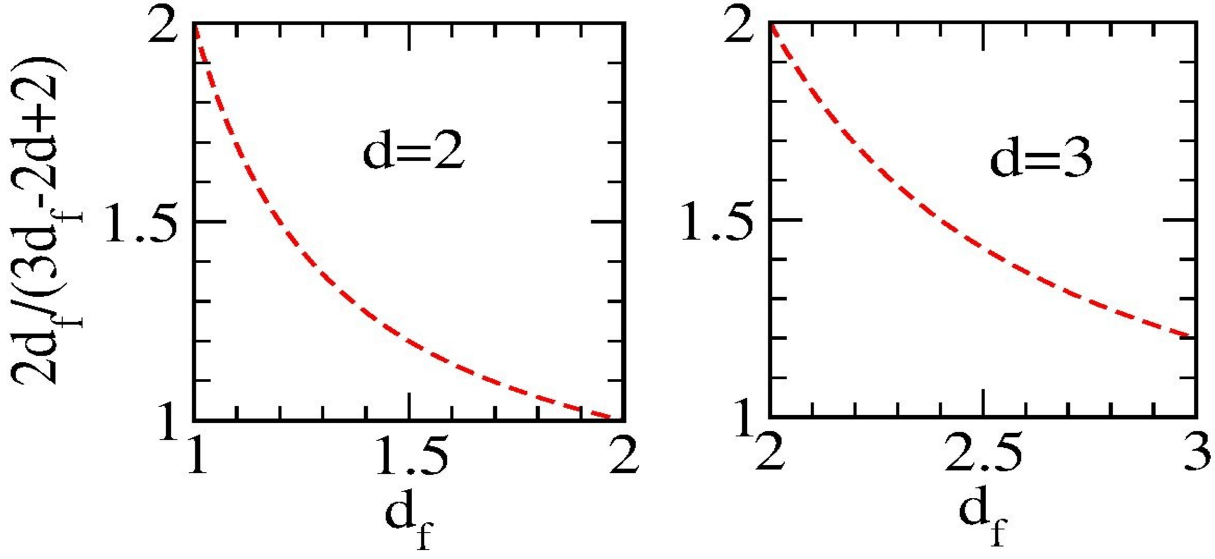
To test the theory, i.e., dependence upon $d_f$,
one can change it by varying temperature (T).




J. Midya and SKD, Phys. Rev. Lett. **118**, 165701 (2017).

In d dimensions: collision cross section $R_g^{d-1} = M^{\frac{d-1}{d_f}}$

$$M \sim t^{\beta}; \quad \beta = 2d_f/(3d_f - 2d + 2)$$



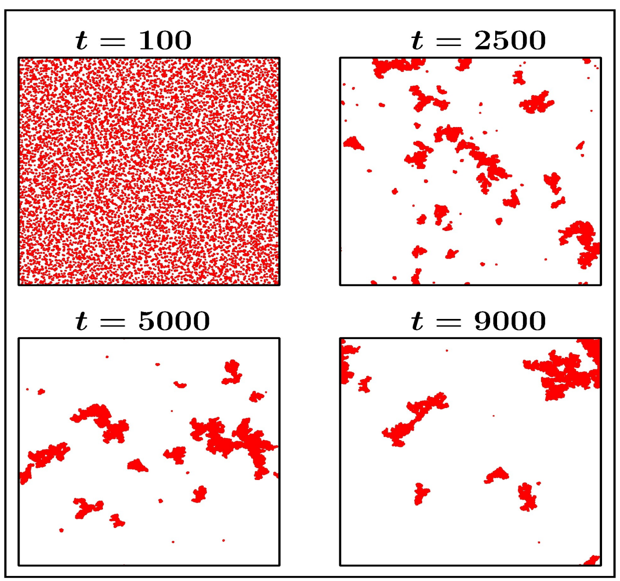

2D: Ballistic aggregation exponent is same as Binder-Stauffer exponent for $d_f = 2$.

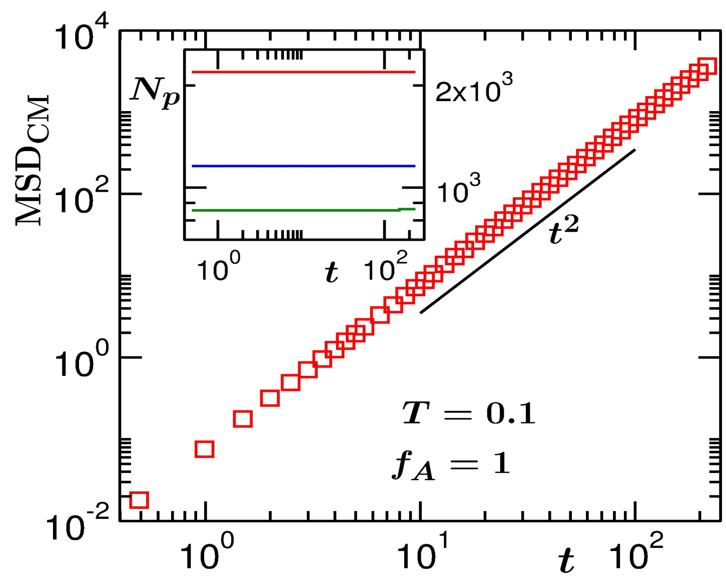
3D: Possibility of exponential growth for extremely filament like structure with $d_f = 4/3$. But not a physical picture.

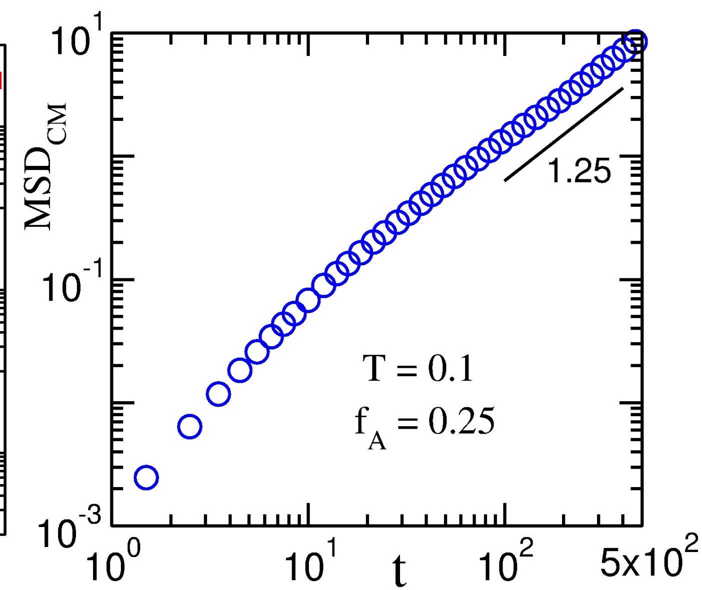
# Phase separation in a Vicsek-like Active matter model

$$m\frac{d^2\vec{r}}{dt^2} = -\vec{\nabla} U_i - \gamma m \frac{d\vec{r}}{dt} + \sqrt{6\gamma k_B T m}\,\vec{R}(t) + \vec{f}_i \qquad U_i \longrightarrow \text{LJ}$$

$$\langle R_{i\mu}(t) R_{j\nu}(t')\rangle = \delta_{ij}\delta\mu\nu\delta(t-t') \qquad \vec{f}_i = f_A \vec{D}_N; \qquad \vec{D}_N = \frac{\sum_j \vec{v}_j}{mag \sum_j \vec{v}_j}$$




By tuning the temperature and active force all types of combination of fractality and cluster mean-squared-displacements can be obtained.

Role of fractality in processes related to diffusive to ballistic coalescence can be understood.

S. Paul and SKD, to be published.

## Summary:

Structure and dynamics related to nucleation and growth in various systems have been discussed.

As opposed to Ostwald ripening mechanism in solid binary mixtures or Binder-Stauffer diffusive coalescence mechanism in liquid-liquid or vapor-liquid transitions, in vapor-solid transition for off-critical density, clusters/droplets grow via ballistic aggregation mechanism.

Similar features have been identified in an active matter model.

*Acknowledgement*: Department of Science and Technology, India
Marie-Curie Actions Plan of European Commission
International Centre for Theoretical Physics, Italy