Clustering of penetrable, athermal active particles that wait on collisions

Bachelorarbeit aus der Physik

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19.03.2024

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Abstract

A system of orientationally persistent, spherical active particles, without thermal or noise effects, that wait when they collide, but can otherwise freely move through each other, is simulated and investigated for clustering behaviour. It is explained how the event-based simulation of this system works. Then, a simple theoretical model is built attempting to describe the system and find conditions for clustering. The simulations show a clustering behaviour that depends on how the waiting time of the particles is determined.

Contents

1 Background

Systems of many interacting, moving particles can show very interesting behaviour, even if the particles follow very simple rules. Often, the particles are passive and obey the classical laws of motion. However, systems of so-called active particles, that are self-propelled (motility) and follow arbitrary rules, can also be considered. Such non-equilibrium systems do not have to conserve energy or momentum. Real world examples, that can be modeled as systems of active particles, are swimming bacteria, birds forming flocks or crowds of humans.

A phenomenon, that active particles can exhibit, is phase separation, where clearly separated regions of high and low particle density (dense and dilute phases) form [1]. Motility induced phase separation (MIPS) can even happen for particles that have purely repulsive interactions where passive particles would rather spread evenly, forming a homogeneous phase [1].

1.1 Microscopic active particle models

Often, one considers Brownian particles, that have Brownian motion and are overdamped, i.e. they have no inertia and the velocity is proportional to the force. This models small particles, suspended in a fluid, with a mass so small and hydrodynamic forces so large that inertia can be neglected. The Brownian motion comes from random impacts of the fluid's particles having thermal kinetic energy. These impacts are modeled by a white, thermal noise F_T in the force. Active particles have a force F_a that represents their active motion, like a bacterium actively swimming. So for the velocity of a particle i we have the overdamped Langevin equation

$$\boldsymbol{v}_i = \frac{1}{\gamma}(\boldsymbol{F}_{\mathrm{a},i} + \boldsymbol{F}_{\mathrm{int},i} + \boldsymbol{F}_{\mathrm{T},i}) \tag{1}$$

with the force $\boldsymbol{F}_{\mathrm{int},i}$ of interactions with other particles. The thermal noise is such, that

$$\langle \boldsymbol{F}_{\mathrm{T},i}(t)\boldsymbol{F}_{\mathrm{T},j}(t')\rangle = \gamma\sqrt{2D_T}\delta_{ij}\delta(t-t')$$

holds, where D_T is the diffusion constant for the diffusion that such particles would undergo without interaction or active force [2].

In the model of active Brownian particles (ABP), the orientation $\hat{\boldsymbol{v}}_0$ of a particle, i.e. the direction of the active force $\boldsymbol{F}_\mathrm{a} = \gamma v_0 \hat{\boldsymbol{v}}_0$, also diffuses with a rotational diffusion constant D_r [2]. This leads to a persistence time τ of the orientation with $\frac{1}{\tau} \propto D_r$ [2]. The Péclet number $\mathrm{Pe} = \frac{3v_0}{D_r\sigma}$ is a measure for the persistence length relative to the particle diameter σ [2].

The model of run-and-tumble particles (RTP) consists of particles that move in a straight line with some speed, and suddenly change their direction to a random new, independent one at random times [3]. Some bacteria behave in a similar way [3].

Without interactions, the dynamics of both, ABP and RTP, represent a diffusive random walk on a larger scale [3].

More interesting behaviour can be seen in systems where the particles interact. This can, for instance, be in the form of interaction forces or aligning the orientation with nearby particles. Another possibility is changing the speed v_0 of the active motion in the vicinity to other particles, i.e. depending on the local density of particles [2]. This behaviour is an example of quorum sensing, seen in some bacteria [2].

1.2 Motility induced phase separation

Consider a system of active particles, with an effective speed that depends on the local particle density. This may be due to quorum sensing or other interactions between particles. For example, they could be slowing down by being in each others way with repulsive interaction.

If there are places where the particles move slower than elsewhere, they spend more time there, as it takes them longer to leave such regions [2]. Thereby, the density of particles is higher there[2]. With the particles' speed v depending on the local density ρ, this can create a feedback loop by the following argument, slightly modified from [3]. With no translational diffusion $\boldsymbol{F}_T = 0$, for the steady-state density $\rho_s = \frac{c}{v}$ being inversely proportional to the speed and v depending on ρ, we get

$$\frac{\mathrm{d}\rho_s}{\mathrm{d}\rho} = \frac{\partial\rho_s}{\partial v}\frac{\mathrm{d}v}{\mathrm{d}\rho} = -\frac{c}{v^2}\frac{\mathrm{d}v}{\mathrm{d}\rho} = -\frac{\rho_s}{v}\frac{\mathrm{d}v}{\mathrm{d}\rho}$$

for how the steady-state density is affected by variations of the density. If this is larger than one, i.e.

$$\frac{1}{v}\frac{\mathrm{d}v}{\mathrm{d}\rho} < -\frac{1}{\rho_s},$$

a perturbation in the density is amplified by the steady state, forming a positive feedback loop. This instability results in a phase separation by spinodal decomposition, where particles accumulate to a dense phase, leaving a dilute phase behind [2].

With some assumptions and approximations, such systems can be described using free energy, like for equilibrium systems [3].

1.3 Simulation approaches

Motility induced phase separation was investigated by numerical simulations for a variety of different systems, for example spherical particles with repulsive interactions with [4] and without [1] translational thermal noise and for infinite orientational persistence length [5].

In these systems, interactions between particles (overlapping with a repulsive force or sliding along each other's surface) last for non-zero time. They can't be described as singular events at certain points in time, as a particle can have multiple interactions with others at the same time, affecting each other. The interaction process can thus be arbitrarily complicated. Therefore, this kind of molecular dynamics simulations requires numerically integrating equation (1) for all particles in small time steps.

Dynamics, that consist of singular events at single points in time, with easily predictable behaviour (like particles moving in straight lines at constant speed) in the meantime, can be simulated using an event based approach. The simulation is then done by processing one event after the other, each time determining the next one.

2 System dynamics

Here, a three dimensional system of active particles is investigated, that can freely overlap without interaction forces, but have to wait for a certain time when colliding with another one. All particles are spherical, have the same diameter σ and move with the same speed v, each in an individual direction, or don't move, when they are waiting. There is no translational or orientational noise, corresponding to zero temperature and infinite persistence length. The distribution of orientations over all particles is isotropic, i.e. there is no globally preferred direction.

The waiting on collisions in this system somewhat resembles the mutual obstruction of particles that happens in the systems mentioned in section 1.3, but can be simulated with an event based approach.

Conditions for phase separation in this system are sought. When referring to particles, their center points are usually meant in the following.

The time T a particle has to wait after a collision, before being released and continuing its motion, is determined when the collision happens. It is the sum of a constant T_0, equal for all particles, and an additional time that depends on the local number density of particles ρ at the time of the collision. Specifically, the additional time is proportional to the number $N_{5\sigma^3} - 2$ of particles within a spherical volume of $5\sigma^3$ around the particle, excluding the particle and its collision partner themselves. The volume is slightly larger than necessary to include particles from previous collisions that are just touching the particle, being at a distance of σ.

$$T = T_0 + T_{\mathrm{d}\rho}(N_{5\sigma^3} - 2) \tag{2}$$

When two particles collide, they stop and wait for at least the time T determined for each particle as just described. If a particle collides with a waiting, still one, T is newly determined for the already waiting particle. If it is larger than the previously remaining waiting time, the wait is extended such that T is the new remaining waiting time, otherwise the old release time remains.

A collision is defined here as the event where the distance of two particles, that are getting closer, reaches one particle diameter. Particles that already overlap do not collide. A particle can only collide with another particle it has collided with before, if at least one of them has moved in the meantime. Otherwise the particles just pass through each other.

For example, consider $T = T_0$ for simplicity and two moving particles A and B that collide at time t_{AB}. They stop moving immediately at t_{AB}. If no other particle is near, A and B will both start moving again at $t_{AB} + T$ and pass through each other. If, instead, a third particle C collides with A at t_{AC} with $t_{AB} < t_{AC} \leq t_{AB} + T$, C stops too. At $t_{AB} + T$ only B is released and starts moving (through A). If B does not collide with C, A and C both start moving at $t_{AC} + T$.

The system was simulated as described in section 4, with results presented in section 5, and modelled for theoretical considerations in section 3.

σ	Diameter of the particles.
T	Minimum time a particle waits after a collision. Can differ between particles and collisions, see equation (2).
T_0	Constant part of T, see equation (2).
$T_{\mathrm{d}\rho}$	Additional waiting time per nearby particle, see equation (2).
T'	Actual waiting time from stopping (first collision) to release of a particle.
a	"Activity". Local fraction of particles that move.
v	The speed of a free particle.
ρ	Local number density of particles.
ρ_a	Local number density of moving particles, $\rho_a = a\rho$.
j	Flux of particles passing from one region to another region, separated by a boundary area.
k	Proportionality constant relating the number of stopping collisions to the density, see equation (4).
T_{delay}	Time delay, compared to free motion, a particle gets due to collisions when travelling a certain distance d, see equation (5).
ρ'	Constant density scale used in the assumptions for $\langle T' \rangle$ in sections 3.2.1 and 3.2.2.
q	Constant exponent used in the assumptions for $\langle T' \rangle$ in sections 3.2.1 and 3.2.2.
λ	Rate of collisions a waiting particle experiences, see equation (9).
g	Proportionality constant relating λ to the density of moving particles $a\rho$, see equation (9).
D	Function $D(a, \rho)$ for the time delay per travelled distance T_{delay}/d, see equation (11).

Table 1: Overview over some of the symbols used in section 3 with their meaning

3 Theoretic modelling

First, we try to model the system described in section 2 to get an understanding of when particles can or cannot be expected to form clusters. Table 1 gives an overview over some of the symbols that were or will be introduced.

3.1 Assumption about flux

Consider a spherical, homogeneous cluster of particles (dense region) of radius r_c in which particles have an effective speed $v_c = av$, where the activity a is the fraction of particles that are moving and not waiting. A particle hitting the cluster has, on average, to travel a distance of

$$\frac{\frac{4}{3}\pi r_c^3}{\pi r_c^2} = \frac{4}{3} r_c \,,$$

which takes it a time of $t_c = \frac{4}{3}\frac{r_c}{v_c}$, which is the average time a particle is part of the cluster. In steady state, the rate of particles leaving the cluster is $\frac{N}{t_c}$, where N is the number of particles in the cluster. With the cluster's density ρ and surface area A_c, the mean flux of particles leaving the cluster

$$j = \frac{N}{t_c A_c} = \frac{\frac{4}{3}\pi r_c^3 \rho}{\frac{4}{3}\frac{r_c}{v_c} \cdot 4\pi r_c^2} = \frac{1}{4}\rho v_c = \frac{1}{4}\rho_a v \,,$$

is proportional to ρ_a and v.

We assume in general, for the flux j of particles passing through an area, from a side with density ρ, activity a and effective speed v_c to the other side, that

$$j \propto v_c \rho = va\rho = v\rho_a \,. \tag{3}$$

This is regardless of what is on the other side or of particles passing through the area in the opposite direction. It means that in a steady state, the density of moving particles $a\rho$ must be uniform.

The assumption ignores that the distribution of orientations of the moving particles within a region might not be isotropic, as it is globally.

3.2 Effects of the waiting time

When travelling a distance d, the front surface of a particle passes through a volume proportional to d (or rather the volume where other particles can be, such that the particle collides with them, is proportional to d). Therefore, it is expected to collide with a number n_{stops} of particles proportional to ρd while moving along its way.

$$n_{\text{stops}} = k\rho d \tag{4}$$

with a proportionality constant k. On each of these collisions, the particle stops for at least a time T. During the waiting time, however, additional collisions can occur with surrounding moving particles. Each additional collision could extend the waiting time. If the average time the particle waits after stopping is $\langle T' \rangle$, the total time delay due to waiting on on its way is

$$T_{\text{delay}} = n_{\text{stops}} \langle T' \rangle = k\rho d \langle T' \rangle \ . \tag{5}$$

The activity a is also the time that a particle moves when passing the distance d, divided by the total time it takes. The total time is the time the particle moves plus the time T_{delay} that it waits.

$$a = \frac{\frac{d}{v}}{\frac{d}{v} + T_{\text{delay}}} = \frac{1}{1 + vk\rho \langle T' \rangle} \tag{6}$$

Consider a boundary area separating two regions of different densities ρ_1 and ρ_2 and activities a_1 and a_2 and the fluxes j_1 and j_2 of particles passing from region 1 to region 2 and vice versa. If the flux of particles going from the region of higher density to that of lower density is larger than the opposite flux, this means that particles spread out, reducing the difference of densities. If this continues, a stable homogeneous phase forms. If, on the other hand, the net particle flux goes from the region of lower density to that of higher, this increases the difference in densities even further. Starting from a homogeneous phase, this is an instability leading to the formation of two coexisting phases. If this continues, particles accumulate to small clusters of high density.

Let's now see what happens for different assumptions about the waiting time $\langle T' \rangle$. In sections 3.2.1 and 3.2.2 we make arbitrary assumptions about $\langle T' \rangle$ to explore the system's dependence on it, before we attempt to estimate the real waiting time in section 3.2.3.

3.2.1 Waiting time as power law of density

If $\langle T' \rangle = T_0 \left(\frac{\rho}{\rho'}\right)^q$ for some constants ρ' and q, equation 3 for the flux from region n becomes

$$j_n \propto v \frac{\rho_n}{1 + vk\rho_n T_0 \left(\frac{\rho_n}{\rho'}\right)^q} \ .$$

We choose $\rho_1 > \rho_2$. Then, $j_1 < j_2$ means that more particles go from the region of lower density to the region of higher density than in the other direction, so we are in the regime of coexisting phases. Let's find the condition for that:

$$\begin{aligned}
& j_1 < j_2 \\
\Leftrightarrow\ & Av \frac{\rho_1}{1 + vk\rho_1 T_0 \left(\frac{\rho_1}{\rho'}\right)^q} < Av \frac{\rho_2}{1 + vk\rho_2 T_0 \left(\frac{\rho_2}{\rho'}\right)^q} \\
\Leftrightarrow\ & \rho_1 \left(1 + vk\rho_2 T_0 \left(\frac{\rho_2}{\rho'}\right)^q\right) < \rho_2 \left(1 + vk\rho_1 T_0 \left(\frac{\rho_1}{\rho'}\right)^q\right) \\
\Leftrightarrow\ & \rho_1 \left(\rho'^q + vkT_0 \rho_2^{q+1}\right) < \rho_2 \left(\rho'^q + vkT_0 \rho_1^{q+1}\right) \\
\Leftrightarrow\ & \rho'^q (\rho_1 - \rho_2) < vkT_0 \left(\rho_2 \rho_1^{q+1} - \rho_1 \rho_2^{q+1}\right) \\
\Leftrightarrow\ & \frac{\rho'^q}{vkT_0} < \frac{\rho_1^q - \rho_2^q}{\rho_1 - \rho_2} \rho_1 \rho_2
\end{aligned} \tag{7}$$

If the average waiting time $\langle T' \rangle$ is independent of ρ, i.e. $q = 0$, the right-hand side is zero, so we are always in the spreading regime and expect a single homogeneous phase. For our system, this would correspond to the case that the waiting time does not depend on the density and could not be extended by further collisions during waiting.

If $\langle T' \rangle$ is proportional to the density, i.e. $q = 1$, the right-hand side becomes $\rho_1 \rho_2$. Then, the coexistence regime is where the product $\rho_1 \rho_2$ is above the threshold of $\frac{\rho'^q}{vkT_0}$. The spreading regime is where the product is below that threshold.

3.2.2 Waiting time as power law of density of moving particles

Assuming $\langle T' \rangle = T_0 \left(\frac{a\rho}{\rho'}\right)^q$ for some constants ρ' and q, equation (6) becomes

$$\begin{aligned} a &= \frac{1}{1 + vk\rho T_0 \left(\frac{a\rho}{\rho'}\right)^q} \\ \Leftrightarrow a + \frac{vkT_0}{\rho'^q} (a\rho)^{q+1} &= 1 \\ \Leftrightarrow a = 1 - \frac{vkT_0}{\rho'^q} (a\rho)^{q+1} &\ . \end{aligned} \tag{8}$$

For $q > -1$ the right-hand side of equation (8) is strictly decreasing in $a\rho$. A higher $a\rho$ then corresponds to a smaller a and thereby a higher ρ. For adjacent regions with densities ρ_1 and ρ_2 and $\rho_1 > \rho_2$, this means that also $a_1\rho_1 > a_2\rho_2$, so, by equation 3, more particles will go from region 1 to region 2 than in the other direction. We would therefore expect such a system to have a single, homogeneous phase.

3.2.3 Deductive model

We now assume that the collisions that happen to a waiting particle, extending its waiting time, occur stochastically and uniformly (as a Poisson process) with a rate λ proportional to the density of moving particles.

$$\lambda = \frac{g}{v} \rho_a v = ga\rho \tag{9}$$

with $\frac{g}{v}$ as proportionality constant.

In contrast to the arbitrary previous assumptions for $\langle T' \rangle$, this is consistent with equation 3 when considering λ as the rate of particles that would pass through the surface of the particle (or rather its interaction range).

Algorithm 1 Algorithm for sampling the distribution of T' and $\langle T' \rangle$

1: Considering a particle that has just stopped due to a collision, set T' to zero as no waiting time has passed yet.
2: Draw a random number t from the exponential probability distribution $\lambda e^{-\lambda t}$, representing the time until the next collision of the waiting particle.
3: **if** $t \leq T$, i.e. the next collision happens before the particle stops waiting, **then**
4: Add t to T', as the next collision extends the particle's waiting time by that amount.
5: Repeat from line 2 with drawing the next number.
6: **else if** $t > T$, i.e. no further collision occurs within the waiting time, **then**
7: Add the remaining waiting time T to T'.
8: Remember T' and repeat from the start until enough samples for T' are found.
9: **end if**
10: When enough samples for T' are found, calculate their mean $\langle T' \rangle$.
11: Repeat everything for different T and/or λ to find how $\langle T' \rangle$ depends on it.

The probability distribution of the waiting time T' and its average $\langle T' \rangle$ resulting from this assumption was numerically sampled equivalently to algorithm 1. Figure 1 shows the sampled distribution for T' with $\lambda T = 1.5$. In the sampled range from $\lambda T = 0$ to $\lambda T = 8$, the values found

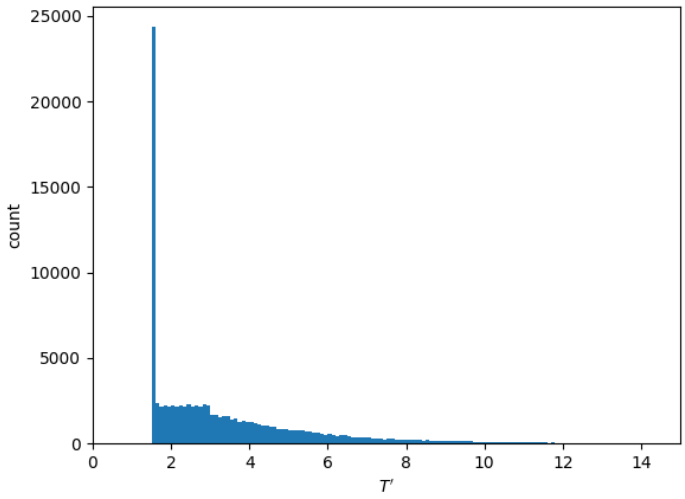


Figure 1: Histogram of the values for T', sampled according to algorithm 1 with $\lambda = 1$, $T = 1.5$ (i.e. there are on average 1.5 collisions per T) and 100000 samples. The bins are of width 0.1 and the histogram is cut off at $T' = 15$. The peak at $T' = 1.5$ corresponds to a Dirac delta in the probability distribution of $\langle T' \rangle$ that represents the finite probability that no collision occurs.

for $\langle T' \rangle$ match $\langle T' \rangle = \frac{1}{\lambda}\left(e^{\lambda T} - 1\right)$ very well. Plugging this into equation (6) gives

$$\begin{aligned} a &= \frac{1}{1 + \frac{kv}{ga}\left(e^{ga\rho T} - 1\right)} \\ \Leftrightarrow a + \frac{kv}{g}\left(e^{a\rho g T} - 1\right) &= 1 \\ \Leftrightarrow a = 1 - \frac{kv}{g}\left(e^{a\rho g T} - 1\right)&. \end{aligned} \tag{10}$$

For $T = T_0$ not depending on density, the right-hand side of equation (10) is strictly decreasing in $a\rho$. Therefore, using the same argument as for equation (8) with $q > -1$, there is not instability and the particles form a homogeneous phase in this case.

3.3 Instability condition

Knowing what to expect for $T = T_0$ from equation (10) or for the waiting times $\langle T' \rangle$ assumed in sections 3.2.1 and 3.2.2, we now derive more general instability conditions. We start with the condition for the delay of particles per distance travelled $\frac{T_{\text{delay}}}{d}$ from equation (6) (because it more general than $\langle T' \rangle$) and consider it a function $D(a, \rho)$ of a and ρ.

$$a = \frac{\frac{d}{v}}{\frac{d}{v} + T_{\text{delay}}} = \frac{1}{1 + v\frac{T_{\text{delay}}}{d}} = \frac{1}{1 + vD(a,\rho)} \tag{11}$$

$$\Leftrightarrow a = 1 - avD(a,\rho) \tag{12}$$

A homogeneous phase is unstable, if small differences in density cause a flux of particles amplifying the differences. Because, by equation 3, the flux of particles leaving a region is proportional to $a\rho$, an amplifying flux arises if $a\rho$ is smaller in the region of higher density ρ. In the limit of a small difference in density, this means

$$\begin{aligned} \frac{\mathrm{d}}{\mathrm{d}\rho} a\rho &< 0 \\ \Leftrightarrow a + \rho\frac{\mathrm{d}a}{\mathrm{d}\rho} &< 0 \\ \Leftrightarrow \frac{\mathrm{d}a}{\mathrm{d}\rho} &< -\frac{a}{\rho}\,. \end{aligned} \tag{13}$$

Considering a as a function, given in equation (12),

$$\frac{\mathrm{d}a}{\mathrm{d}\rho} = \frac{\partial a}{\partial \rho} + \frac{\partial a}{\partial a}\frac{\mathrm{d}a}{\mathrm{d}\rho}$$
$$\Leftrightarrow \frac{\mathrm{d}a}{\mathrm{d}\rho} = \frac{1}{1-\frac{\partial a}{\partial a}}\frac{\partial a}{\partial \rho} = \frac{-av}{1+vD+av\frac{\partial D}{\partial a}}\frac{\partial D}{\partial \rho}\,.$$

So equation (13) becomes

$$\frac{-av}{1+vD+av\frac{\partial D}{\partial a}}\frac{\partial D}{\partial \rho} < -\frac{a}{\rho}$$
$$\Leftrightarrow \frac{\rho}{\frac{1}{v}+D+a\frac{\partial D}{\partial a}}\frac{\partial D}{\partial \rho} > 1\,.$$

With $D = k\rho\,\langle T'\rangle$ as before, this becomes

$$\frac{\rho}{\frac{1}{v}+k\rho\,\langle T'\rangle + ka\rho\frac{\partial\langle T'\rangle}{\partial a}}k\left(\langle T'\rangle + \rho\frac{\partial\,\langle T'\rangle}{\partial\rho}\right) > 1\,. \tag{14}$$

If the denominator is negative, $\frac{\partial\langle T'\rangle}{\partial\rho}$ also needs to be negative to fulfill this. If that is not the case, we get

$$\rho k\left(\langle T'\rangle + \rho\frac{\partial\,\langle T'\rangle}{\partial\rho}\right) > \frac{1}{v} + k\rho\,\langle T'\rangle + ka\rho\frac{\partial\,\langle T'\rangle}{\partial a}$$
$$\Leftrightarrow \rho\left(\rho\frac{\partial\,\langle T'\rangle}{\partial\rho} - a\frac{\partial\,\langle T'\rangle}{\partial a}\right) > \frac{1}{vk}\,.$$

Note, that the left-hand side vanishes if $\langle T'\rangle\,(a,\rho)$ is a function of $a\rho$, so the system can't be unstable then. This is the case in section 3.2.2 and in section 3.2.3 for constant T.

In section 3.2.1 we had $\langle T'\rangle = T_0\left(\frac{\rho}{\rho'}\right)^q$, then the condition becomes,

$$q\rho^{q+1} > \frac{\rho'^q}{vkT_0}\,.$$

This is equation (7) in the limit of $\rho_2 \to \rho_1 = \rho$.

Taking the deductive model from section 3.2.3 with $\langle T'\rangle = \frac{1}{ga\rho}\left(e^{ga\rho T}-1\right)$ and letting T be a function $T(a,\rho)$ of a and ρ, we have

$$\frac{\partial\,\langle T'\rangle}{\partial a} = -\frac{\langle T'\rangle}{a} + \frac{1}{a}\left(T + a\frac{\partial T}{\partial a}\right)e^{ga\rho T}$$

and similarly $\frac{\partial\langle T'\rangle}{\partial\rho}$, so the condition becomes

$$\rho\left(-\langle T'\rangle + \left(T+\rho\frac{\partial T}{\partial\rho}\right)e^{ga\rho T} + \langle T'\rangle - \left(T+a\frac{\partial T}{\partial a}\right)e^{ga\rho T}\right) > \frac{1}{vk}$$
$$\Leftrightarrow \rho e^{ga\rho T}\left(\rho\frac{\partial T}{\partial\rho} - a\frac{\partial T}{\partial a}\right) > \frac{1}{vk}\,.$$

So for constant T the system cannot be unstable. If, however, T increases with the density ρ or decreases with the activity a (as long as the denominator in equation (14) stays positive), the system can become unstable for sufficiently large ρ and a.

With T being determined by equation (2), we get

$$\rho^2 e^{ga\rho T}\frac{T_{\mathrm{d}\rho}}{5\sigma^3} > \frac{1}{vk}$$

so we expect an instability for sufficiently large ρ and $T_{\mathrm{d}\rho}$.

4 Implementation of the simulation

A simulation of the system described in section 2 in a cubic box with periodic boundary conditions was implemented in C++ with an event based approach. The idea is, that events, like collisions, are scheduled for a certain time. In a loop, the earliest event is selected and the system's state changed accordingly. If the handled event causes new events, they are scheduled.

To find the next collision of a particle, it must be checked for collisions with every other particle. As this must be done regularly for each particle, the effort for that scales with the square of the number of particles. To reduce it, the simulation box is split into cubic zones with a side length slightly larger than the particle's diameter. With that, as long as no particle switches zones, particles can only collide with particles in the same zone as the particle itself or in neighboring zones, greatly reducing the number of required checks. When a particle does switch zones, this is considered an event that triggers the calculation of possible collisions of that particle for the new zone. The zones are also large enough to find all particles, that need to be counted to determine the waiting time according to equation (2), in the same or neighboring zones.

Particles and events are represented by objects. There are three types of events: collisions, releases (when a particle stops waiting) and zone-switches. A special, fourth type of event is used too, as mentioned later. Each event object stores the time of the event and holds references to the involved particle(s). Scheduled events are kept in a queue, that allows access to the earliest of the events it contains. The next event is handled by removing it from the queue and calling the corresponding handler-method of the involved particle(s).

Not all scheduled events actually happen. An event can be invalidated if it is affected by a previous event. For example, consider two particles on colliding trajectories and an event scheduled for their collision. A nearby third particle could be released from waiting in the meantime and take a trajectory that makes it collide with one of the particles before. The originally scheduled collision event will then become outdated. If a particle reaches a collision event that was outdated due to the other particle, it will just find and schedule the next event. Outdated release or zone-switch events and collision events reached by none of the particles do nothing when they are handled.

4.1 Event finding, scheduling and handling

Particles, that have collided and have not moved since then, should not immediately collide again, when being released. But their distance is just one particle diameter, so it would be up to numerical imprecision, whether the particles would collide again or be considered overlapping and not collide. Therefore, each particle keeps track of the particles it collided with, since it last moved. Each waiting particle also holds a reference to a release event and each moving particle one to the next collision or zone-switch event. To find the next event for a particle, possible collisions with the other particles in the same or neighboring zones are calculated based on the current trajectories. Collisions between particles, that both have each other in their sets of particles they collided with, are not taken into account. Neither are those, that happen after one of the particles' referenced events. If the particle is moving, the time at which it would leave the zone is calculated too. The earliest of the events found this way is scheduled. To schedule an event, a corresponding object with the time of the event and the involved particle(s) is inserted into the event queue.

If a particle is moving at the end of handling an (release, zone-switch or outdated) event, it finds and schedules the next event and saves a reference to the it. If the next event is a collision and the collision partner is moving too, that particle also saves a reference and marks its previously referenced event as outdated.

A particle handling a non-outdated collision event adds the collision partner to the set of particles it saves to avoid colliding with particles, that it should pass through, on release. It also calculates the time at which it will be released if no further collisions occur, using equation (2) and such, that the release time never decreases. If the particle was previously moving or had an earlier release event, a release event for the new release time is scheduled and referenced. The possible previously referenced, earlier release event is marked outdated. The particle then looks for collisions before the new release time. If it finds one, it schedules the earliest and the collisions partner references it and marks its previously referenced event outdated.

A waiting particle handling an outdated collision event looks for the next collision before its release, if the outdated event was previously scheduled by itself. Outdated collisions scheduled by other particles do not affect the event scheduled by this particle, so nothing needs to be done.

After a particle is released, its set of particles it collided with since last moving must be cleared, so it can later collide with them again. If two particles collide and then get released at the same time, one of the release events must be handled first. If the first particle's handler would then clear

the set, the other particle would consider the first particle as one it can collide with again, possibly detecting and scheduling an immediate collision erroneously. Instead, the handler schedules another type of event for the same time as the release, which gets handled only after all other events at the same time are handled. This event then clears the set (and does nothing else). Clearing it on the next collision or zone-switch, instead, does not work, because particles that collided with each other can collide again, directly after their release, if they are released at different times. If only one particle having the other one in its set would suffice to prevent a collision, the second collision of a particle with another one, that didn't move in the meantime, would be missed. This can happen due to the periodic boundary conditions.

4.2 Technical details

Each particle saves the time of its last event and the coordinates it had there. At the beginning of handling an event, they are updated to the time of that event. Zone-switch events are not scheduled for a time as close to the actual crossing of the zone's boundary as possible, because, with numerical errors, the particle could still find itself in the old zone when handling the event. Instead, zone-switch events are scheduled for a time where the particle is guaranteed to find itself having left the old zone when handling the event, even with numerical errors. If a particle finds itself in a new zone when handling an event, it switches to the new zone, even if the event is not a zone-switch event. The zones are large enough, so that no collisions can be missed due to a particle overshooting the zone's boundary before switching.

To avoid memory allocations during the simulation, sufficiently many event objects are constructed at initialization and organized in a stack structure where the they are taken from, before being scheduled, and returned to, after being handled.

Coordinates and times are saved using 64-bit integers, because they provide equal absolute precision over the full range. With a floating point representation, not the full range of values could be used and their absolute precision is exponentially reduced with the absolute value of the stored number. Vectors (velocities and differences of coordinates) are represented using `doubles`, because multiplicative calculations have to be done on them.

To use the full available 64-bit range for the coordinates, the size of the particles is scaled to fit the desired volume density η (number of particles times the volume of a particle divided by the volume of the simulation box). The speed of the particles is chosen such, that the simulated time, where the range of integer representation of the time ends, is sufficiently large. The zones have a side length slightly larger than the particles' diameter and such, that they divide the simulation box into (almost) equal parts. Simulations can be performed for up to three dimensional space for arbitrary numbers of particles, packing fractions and waiting times T.

The zones are realized as a doubly linked list of particles for each zone together with an array of pointers to the first particle of each list, or null-pointers, for empty zones. From coordinates, the array-index for the corresponding zone can be calculated.

As the queue containing the scheduled particles, the `priority_queue`, provided by the standard library, is used. The set of particles a particle collided with since last moving is stored using the standard library's `unordered_set`. This may, however, not be a good choice for the typically very small number of particles in there.

During the simulation, the sizes and numbers of clusters, and the number of particles in each zone are be written to a file regularly. The coordinates, velocities, cluster association, and moving/waiting state of each particle can be written too.

At the end of a simulation, the relevant state is saved to a file that can be used to continue the simulation later. To get the exact same result from continuing a simulation as one would get from running it longer without interruption, the order of events scheduled for the same time has to be preserved. Otherwise, for example, release events of particles that happen at the same time can be handled in different order, leading to different sequence of later events being scheduled and possibly outdated. This can cause differences, due to limited numerical precision, that are then amplified.

5 Simulation results

The system described in section 2 was simulated as explained in section 4. Simulation runs were performed with $N = 50000$ particles, for different T_0, $T_{\mathrm{d}\rho}$ and global volume densities $\eta = \frac{1}{6}\pi\sigma^3 N/L^3$, with L the side length of the simulation box. Some of the symbols used in this section can be looked up in table 2.

σ	Diameter of the particles.
T	Minimum time a particle waits after a collision. Can differ between particles and collisions, see equation (2).
T_0	Constant part of T, see equation (2).
$T_{\mathrm{d}\rho}$	Additional waiting time per nearby particle, see equation (2).
v	The speed of a free particle.
η	The global volume density, $\eta = \frac{1}{6}\pi\sigma^3 N/L^3$, with N the number of particles and L the side length of the simulation box.
Z_n	Number of zones, where n particles are in the block of $3 \cdot 3 \cdot 3$ zones around that zone.
S	A distance measure, between the distribution of the number of particles per block of 3^3 zones and the Poisson distribution one would have for randomly, uniformly placed particles. See equation (15).

Table 2: Overview over some of the symbols used in section 5 with their meaning

At initialization, the particles were placed randomly within the simulation box, with a uniform probability distribution. The orientations of the particles were drawn from an isotropic probability distribution, independent of the position. In each of the sections 5.1 and 5.2, the initial distribution of particles, relative to the simulation box, and their orientations is equal, as the same seed was used for the simulations presented there.

In the following, *clusters* refer to the smallest sets of particles, such that two particles, that touch or overlap with each other, are part of the same cluster. *Dense clusters* refer to clusters, where the particles are significantly denser than in the surrounding, and whose existence makes the system non-homogeneous. *Clustering* refers to the formation of dense clusters.

For a random, uniform distribution of the particles, like at initialization, the number of particles found in a zone is expected to follow a Poisson distribution. Let Z_n be the number of blocks of $3 \cdot 3 \cdot 3$ zones (introduced in section 4) that contain exactly n particles, Z the total number of zones (which is also the number of different blocks of 3^3 zones) and $P(n)$ the Poisson-distributed probability to find n particles in a block of 3^3 zones, if all particles were randomly and uniformly distributed, like at initialization. The symmetric chi-squared distance [6] between the observed and the Poisson distribution

$$S = \sum_{n=0}^{\infty} \frac{\left(\frac{Z_n}{Z} - P(n)\right)^2}{\frac{Z_n}{Z} + P(n)} \tag{15}$$

is used as a measure of how non-homogeneous the system is. S can't reach values larger than two. It turns out to be suitable to distinguish states with only one homogeneous phase from those, where dense clusters exist. If the particles would spread even more uniformly than random, S would also increase, but this doesn't happen here.

5.1 Density dependent T

If the minimum waiting time T increases with the local density, as is the case for positive $T_{\mathrm{d}\rho}$, section 3.3 predicts that instabilities, leading to the accumulation of particles, could occur. Simulations for different $T_{\mathrm{d}\rho}$ with $T_0 = 0$ were performed. Some simulations with nonzero T_0 appeared to require longer computation and show qualitatively similar results, and are therefore not considered further. Figure 2 shows the simulated parameters, arranged as a phase diagram.

At small $T_{\mathrm{d}\rho}$ and low densities η, the system almost immediately reaches a state of a homogeneous phase and the distance S fluctuates near zero, i.e., the spatial distribution remains almost random.

For larger $T_{\mathrm{d}\rho}$ or η, the system is indeed unstable and shows clustering where spots of high density appear. These often start to form immediately. However, for densities near the boundary of the parameter region where clustering occurs, the system often keeps a homogeneous phase for a while, but then spontaneously forms a dense cluster in a nucleation process.

Homogeneous states and states with dense clusters differ by a large difference in S. In the phase diagram (figure 2) a clear separation of a homogeneous region and an unstable region can

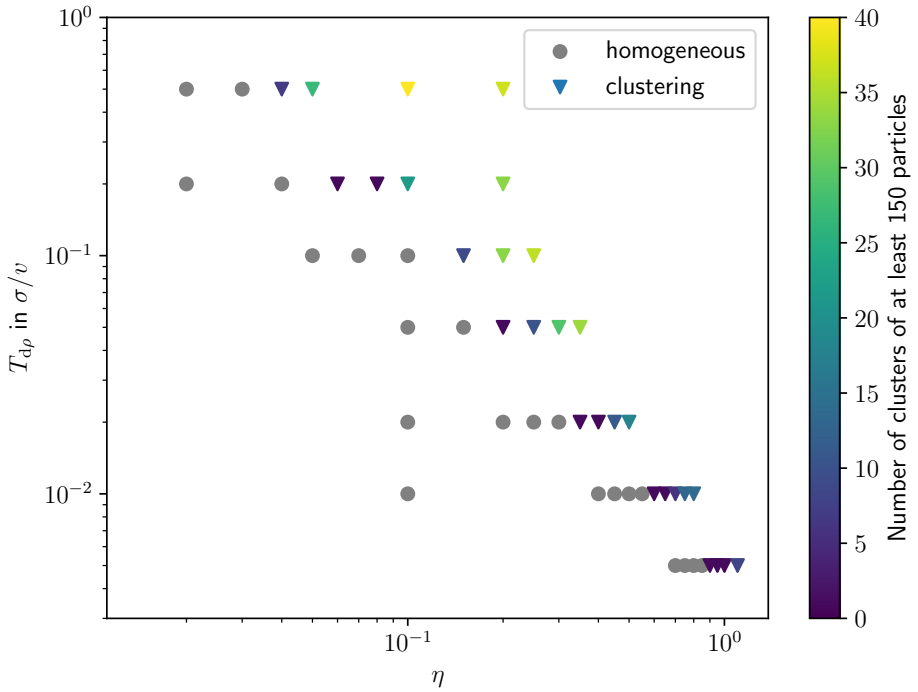


Figure 2: Phase diagram with simulation runs for $T_0 = 0$ with different global volume densities η, and density dependent waiting times $T_{d\rho}$. Runs marked by circles ended with a homogeneous phase with $S < 1$. Those marked by triangles, ended with $S > 1$, indicating that phase separation happened and at least one cluster formed. For the latter, the number of clusters with a size of at least 150 particles is marked by color. Those clusters are larger than the clusters in most homogeneous states (except for the percolating cluster). This number is therefore approximately the number of dense clusters formed as a result of the instability. It tends to decrease close to homogeneous region. Note, that the simulations did not reach a steady state, as mentioned in the main text.

be seen. The boundary appears approximately as a straight line in the double-logarithmic phase diagram.

The spatial size of dense clusters doesn't generally grow, even when growing in particle number, but their density increases instead. Figure 3 shows examples where these behaviours can nicely be seen. The contraction of clusters with increasing density is expected, as the instability leads to an accumulation of particles at more dense places, also within a cluster.

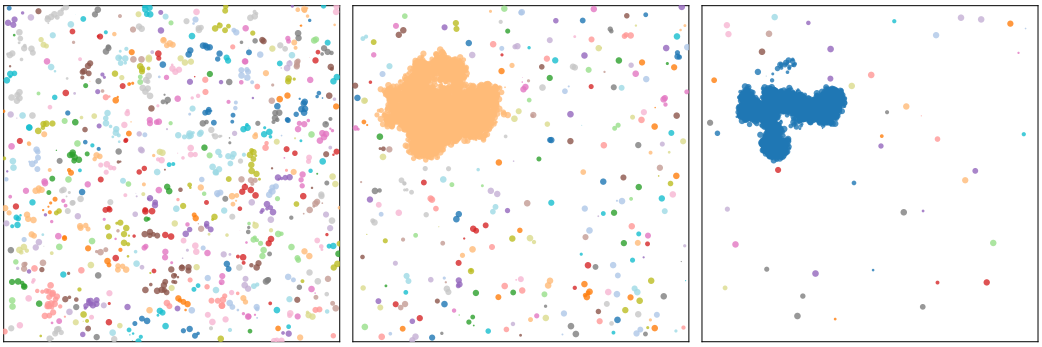
There appears to be a tendency, that the number of dense clusters, that form, becomes smaller when closer to the homogeneous region. This can be seen in figure 2 and also in figure 3. This can be explained by the system being less unstable there, reducing the probability for density fluctuations large enough to trigger the accumulation of more particles, i.e. nucleation, leading to fewer dense clusters. Once dense clusters have formed, they can capture more and more particles, reducing the density in the rest of the system and thereby also reducing the probability of the formation of new dense clusters.

A steady state was not reached within the simulated time in any of the runs where clustering occurred. The number of particles in the most dense block of 3^3 zones kept increasing in all cases. When clustering occurred in a simulation, all simulations for the same $T_{d\rho}$ were continued to at least a time three times as large as the time where the cluster(s) formed. Thus, a simulation ending in a homogeneous phase has maintained that phase for a significant amount of time. Specifically, the simulations ended at a time of $6000\frac{\sigma}{v}$ for $T_{d\rho} = 0.005$, $12000\frac{\sigma}{v}$ for $T_{d\rho} = 0.01$ and $T_{d\rho} = 0.02$, $16000\frac{\sigma}{v}$ for $T_{d\rho} = 0.05$, $40000\frac{\sigma}{v}$ for $T_{d\rho} = 0.1$, $80000\frac{\sigma}{v}$ for $T_{d\rho} = 0.2$ and $160000\frac{\sigma}{v}$ for $T_{d\rho} = 0.5$.

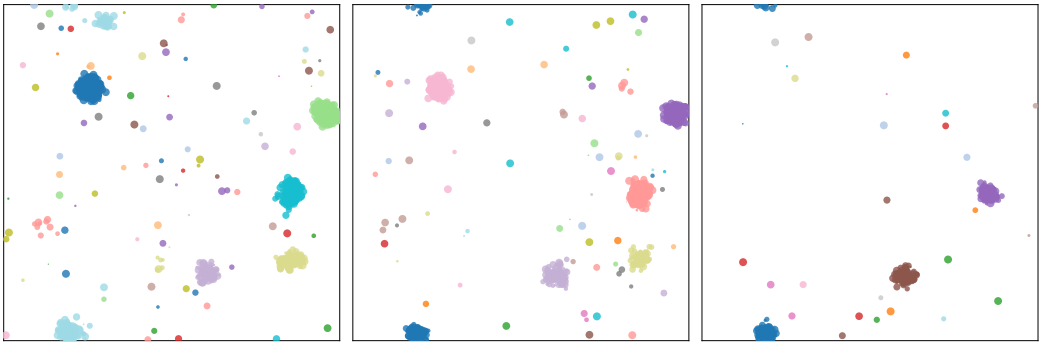
5.2 Constant T

According to the model in section 3, for constant $T = T_0$ the system should always stay as a stable, homogeneous phase. Simulations were performed for $T = T_0$, $T_{d\rho} = 0$ with different parameters that can be seen in figure 4.

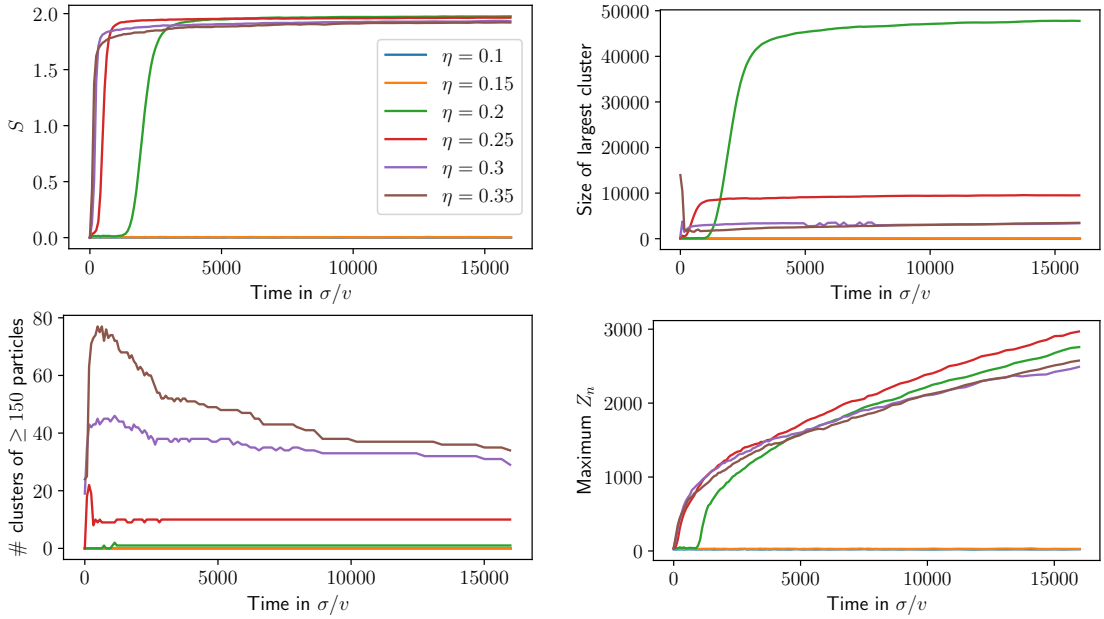
First, we consider the simulations for $T_0 \leq 2\frac{\sigma}{v}$. There, the the system quickly reaches a distribution of cluster sizes that is then maintained up to noise-like fluctuations. The distribution



(a) A plane through the system with $\eta = 0.2$, at times $800\frac{\sigma}{v}$ (left), $2400\frac{\sigma}{v}$ (center) and $16000\frac{\sigma}{v}$ (right), with the intersections of particles with that plane. Particles that belong to the same cluster have the same color (the number of colors is limited, though). The system is near the homogeneous region in the phase diagram and it shows nucleation. On the left, it is still in the homogeneous phase and the decomposition hat not yet started. In the center, a dense cluster has formed and already contains a large fraction of all particles (see top right of figure 3c, green curve). The density everywhere else is reduced, preventing the formation of other dense clusters. On the right, almost all particles are part of the cluster, but the cluster has shrunk spatially, increasing its density (see bottom right of figure 3c).



(b) Plane of the system with $\eta = 0.35$, at times $800\frac{\sigma}{v}$ (left), $2400\frac{\sigma}{v}$ (center) and $16000\frac{\sigma}{v}$ (right). Being farther away from the homogeneous region, multiple dense clusters form immediately, but not all of them survive. See also the brown curves in figure 3c.



(c) Temporal development of S (equation (15)), the size of the largest cluster, the number of dense clusters and the maximum number of particles in a block of 3^3 zones. The legend applies to all of the plots. One can see a delayed decomposition for $\eta = 0.2$ (green), a persistent increase of the highest density, even when the largest cluster doesn't grow anymore, and a decreasing number of dense clusters close to the homogeneous phase (bottom left).

Figure 3: Evolution of the simulated systems with $T_0 = 0$, $T_{\mathrm{d}\rho} = 0.05\frac{\sigma}{v}$.

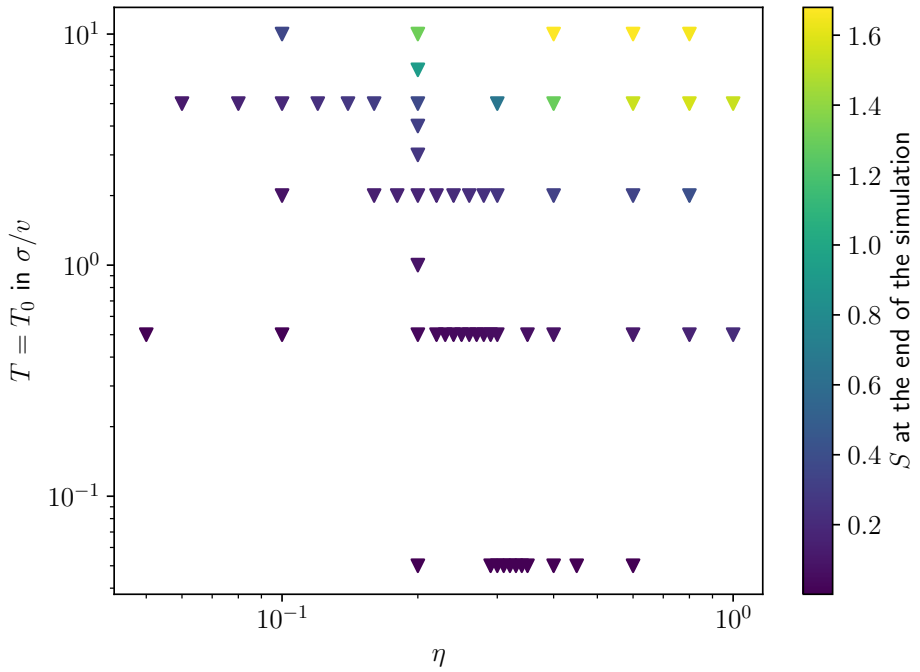


Figure 4: Phase diagram with simulation runs for $T = T_0$, $T_{\mathrm{d}\rho} = 0$ with different global volume densities η, and minimum waiting times T_0. The color denotes the value of S at which the simulations ended. Not all of them reached a steady state, as mentioned in the main text.

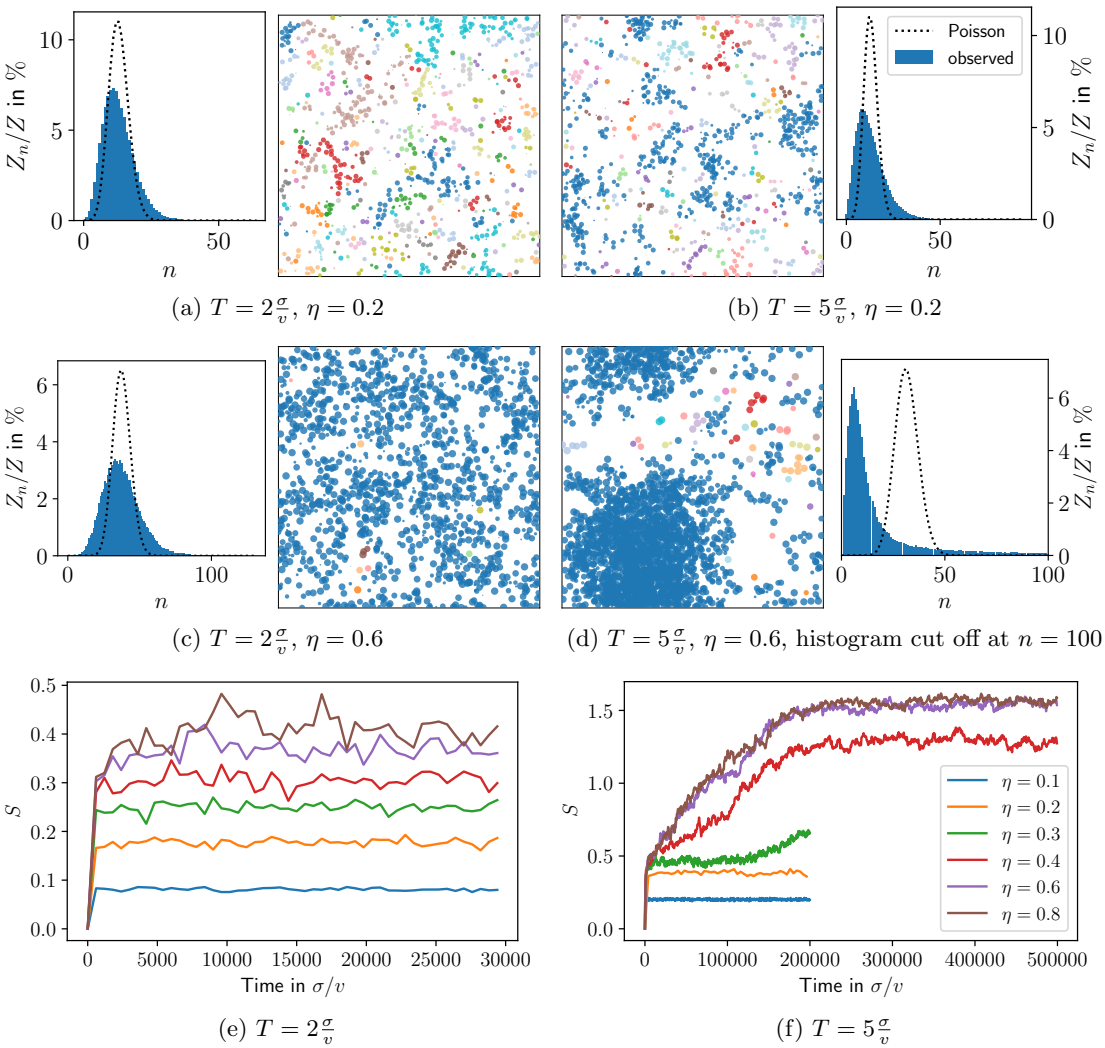
Z_n/Z of numbers of particles ending up in blocks of zones is very close to the Poisson distribution for small T_0 or η and gets wider for larger T_0 and η. So the system is essentially in a homogeneous phase where the distribution of particles gets less uniform with increasing η and T_0. This can be seen in figures 5a to 5c and 5e.

At small densities η, the particles are isolated or form small clusters. As the density increases, larger clusters become more frequent, until the percolation threshold is reached, where a large cluster emerges, that spreads through the entire system. Figure 6 shows that the threshold becomes lower with larger T_0. For the three dimensional Swiss cheese model, a system of randomly placed spheres of equal size, allowed to overlap, like at the initialization of the simulations, the percolation threshold is at about $\eta \approx 0.342$ [7]. This is close to the threshold observed for a small T_0 of $0.05\frac{\sigma}{v}$. The percolation has no effect on the spatial distribution of the particles. Figures 5a and 5c show a system below and above the percolation threshold. This also happens for nonzero $T_{\mathrm{d}\rho}$ but is easier to see here, as no instability is in the region considered here.

For larger $T_0 \geq 0.5\frac{\sigma}{v}$, clustering did occur in the simulations for large enough η. An example can be seen in figure 5d. Figure 5f shows a similar unstable behaviour of S as seen in section 5.1, but much less pronounced and much slower. The distribution of particles in the homogeneous phase is already less uniform and the clusters, that form, are much less dense and their density does not keep increasing. The evolution of S for $T_0 = 5$, $\eta = 0.3$ seen in figure 5f suggests nucleation as also seen in section 5.1. Figure 4 shows by colors the distance S at which the simulations ended. For simulations where S stays below 0.5 appear to be in a steady state, i.e. S and the distribution of cluster sizes only fluctuate a trend. For most cases where S exceeds 0.5, the changing of S and the distribution of cluster sizes stagnated before the end of the simulation but with irregular fluctuations. Not enough simulations were run to tell the shape of the boundary of the unstable behaviour in the phase diagram. The simulations in that region take the longest computation time of those considered here.

6 Discussion

In the case of a positive dependence of the minimum waiting time T on the local density during the collision, the model built in section 3 allows (and expects) an instability that leads to the accumulation of particles and clustering, for sufficiently large densities ρ and density dependencies $T_{\mathrm{d}\rho}$. This is indeed observed in the simulation for positive $T_{\mathrm{d}\rho}$. For constant minimum waiting times $T = T_0$, without a dependence on the density, i.e. $T_{\mathrm{d}\rho} = 0$, the the model is stable and allows only a homogeneous phase. However, in the simulations, an instability leading to the formation of



(a) $T = 2\frac{\sigma}{v}$, $\eta = 0.2$ (b) $T = 5\frac{\sigma}{v}$, $\eta = 0.2$

(c) $T = 2\frac{\sigma}{v}$, $\eta = 0.6$ (d) $T = 5\frac{\sigma}{v}$, $\eta = 0.6$, histogram cut off at $n = 100$

(e) $T = 2\frac{\sigma}{v}$ (f) $T = 5\frac{\sigma}{v}$

Figure 5: Examples for the behaviour of the system with $T_{\mathrm{d}\rho} = 0$ for $T = T_0 = 2\frac{\sigma}{v}$ (left) and $T = T_0 = 5\frac{\sigma}{v}$ (right). The time evolution of S for some densities is shown in (e) and (f) (the legend applies to both). The histograms show the distribution Z_n/Z of numbers of particles n per block of 3^3 zones, so basically the density distribution. The Poisson distribution, that randomly placed particles would have, is marked for comparison. Even for the case with clustering in (d), the distribution has only one peak, so the cluster is in the extended tail. (a)-(c) show a plane through the system with the respective intersections of the particles. Particles that belong to the same cluster have the same color (the number of colors is limited, though). The system in (c) is above the percolation threshold, as compared to (a), so most of the particles belong to the same cluster.

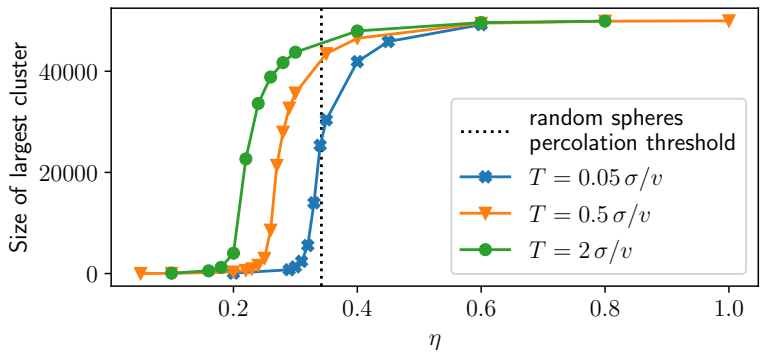


Figure 6: Size of the largest cluster, averaged over 30 points in the time evolution, over the global volume density η for different $T = T_0$. The percolation threshold for a system of randomly places spheres (Swiss cheese model) at $\eta \approx 0.342$ is marked for comparison [7].

clusters is found for very large T_0. So, apparently, at least one of the assumptions that lead to the exclusion of an instability must be wrong. Possibly, the orientation of the particles, which is not taken into account in the model, plays a role. Another possibility is, that the consideration of the system as a density field fails in equation (4) or equation (9).

7 Conclusion

The system of penetrable, orientationally persistent, spherical active particles, without thermal or noise effects, that wait when they collide, was simulated and investigated. A simple theoretical model for the system was built, that is compatible with instabilities found by the simulations for positively density dependent minimum waiting times ($T_{\mathrm{d}\rho} > 0$), but fails to describe instabilities found for large, constant minimum waiting times ($T = T_0$). The instabilities in these two cases were found to be different, in that the density of the formed clusters keeps increasing and the clusters don't grow spatially in one case, but not in the other. The percolation threshold was was found to decrease with increasing T_0. For $T_{\mathrm{d}\rho} > 0$ with $T_0 = 0$, a clear separation of the regions in the η-$T_{\mathrm{d}\rho}$-plane, where clustering does or does not occur, was found. This could be further analysed and compared to models. Also, the instability for large, constant T could be investigated by further simulations, as well as the interplay of $T_{\mathrm{d}\rho} > 0$ and $T_0 > 0$ at the same time.

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Erklärung

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