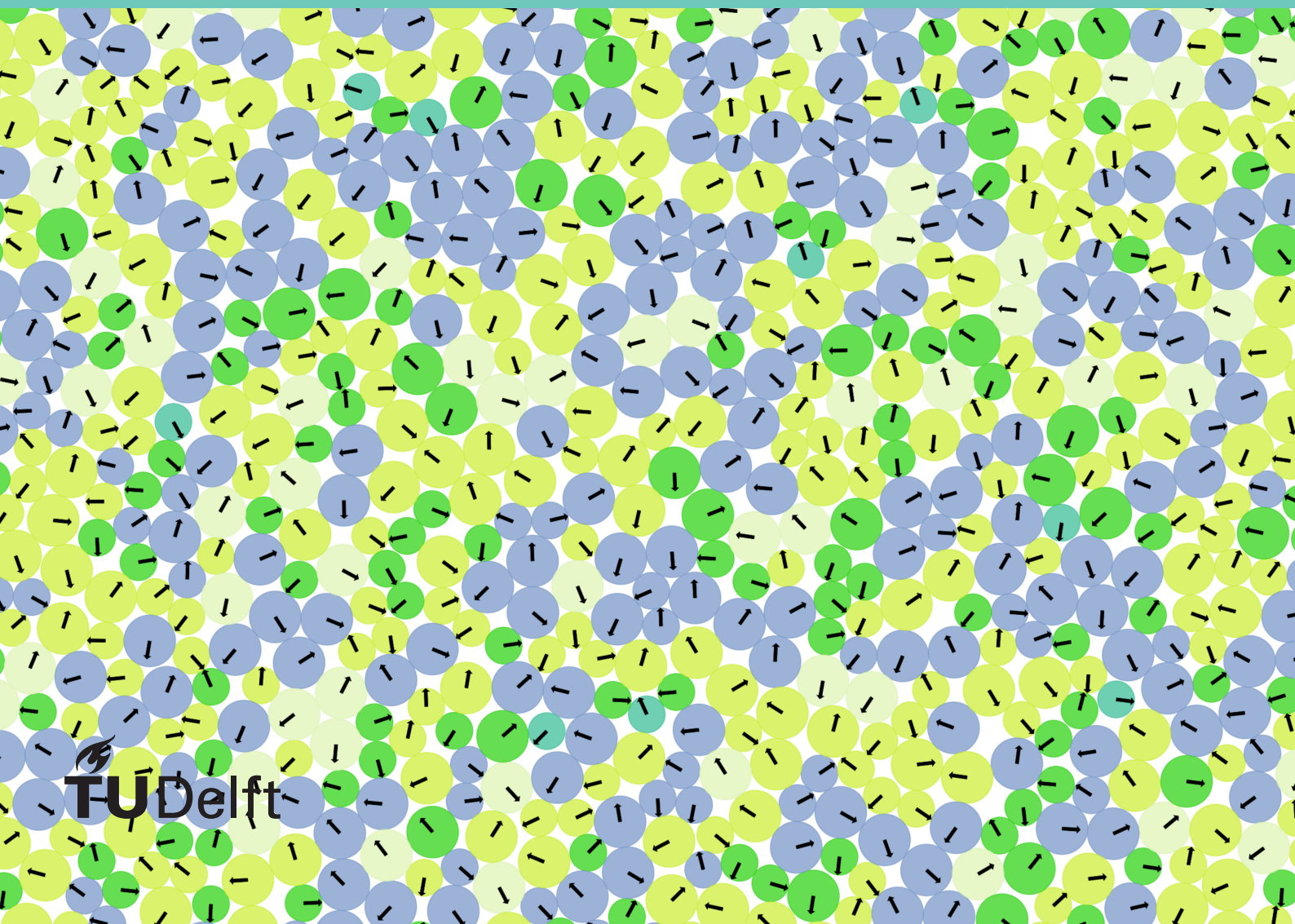


Strain Softening

Abhishek Kumar

Linear and nonlinear response of weakly attractive soft particles to shear strain



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soft particles to shear strain

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Abstract

Emulsions and foams are commonly found in products made by industries ranging from those associated with food and pharmaceuticals to those involved in selling personal care products. This work is motivated by the need for accurate models of their mechanics, which can then be used for efficient processing.

They can be thought of as soft repulsive spheres that can overlap with one another to a certain extent, along with a weakly attractive potential between the spheres. We study such systems in the context of the *jamming transition* - a transition seen in disordered systems from a *flowing* state to one where they *jam* and develop rigidity. The canonical model for the jamming transition is one of soft, repulsive and frictionless spheres which describe many common physical systems. An attractive tail is added to the repulsive potential used in this canonical model, in order to describe systems like emulsions and foams.

We compare the linear response of emulsions and foams with that of the canonical model. Recent studies have shown for the canonical model that when we impose a quasi-static shear strain at the boundaries of disordered systems, the linear elastic regime survives for a small window close to the beginning of the straining action. It gives way to softening in the linear elastic regime, associated with the beginning of a nonlinear response regime. We investigate how this window leading to the non linear response changes for emulsions and foams. The predictions obtained for softening, from ideas that derive from linear response in the jamming transition and by imposing a quasi-static shear strain is compared for both emulsions and the canonical model.

Acknowledgements

It was at my first department borrel that I had met Brian. This was two and a half years ago. As I remember, we had a very interesting conversation on active matter, with Brian doing most of the talking, but never really tiring of the questions I had for him on this subject. When it was time for me to find a guide for my master's thesis work, I remembered the conversation and I knew I wanted to work with Brian. He has been patient with my endless questions through the entirety of my time under his supervision. His encouragement during times when I got stuck with a problem, and his approach to finding solutions to problems instead of dwelling on them have had an immense impact on me and my understanding of the qualities that make an excellent mentor. I'm grateful to have had him as my master's thesis supervisor. Along with him, his Ph. D students Julia and Dion have been a source of a lot of free coffee, interesting conversations and help with the practicalities of doing research in an academic setting.

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List of Symbols

J	Critical point for jamming transition
V_{ij}	Potential between particles i and j in contact
F_{ij}	Force between particles i and j in contact
k_{ij}	Stiffness between particles i and j in contact
δ_{ij}	Relative overlap between particles i and j in contact
ϵ	Energy scaling factor
T	Temperature
Z	Average contact number for particles of a packing
ϕ	Volume fraction of particles in a packing
σ	Shear stress
p	Pressure
B	Bulk Modulus
V	Volume of a packing
L	Length of a packing
G	Shear modulus
a	Attraction strength
γ	Shear strain
R_i	Radius of particle i
r_{ij}	Distance between the centres of particles i and j
U	Potential energy
$\mathbf{K}$	Dynamical matrix
$\mathbf{q}$	Degrees of freedom in a packing
$u_{\parallel}$	Relative displacement parallel to the line of contact between particles in contact
$u_{\perp}$	Relative displacement perpendicular to the line of contact between particles in contact
C	Correlation of particle velocities with their initial velocities at a strain step
γ_s	Strain at which softening is observed for σ/γ
$\gamma_s^{\dagger}$	Strain at which softening is observed in the correlation C

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1

Jamming of repulsive particles

1.1. Motivation for the present work

Glass has a wide variety of uses in modern society. From art to industry, it is an ubiquitous material. What we lack though, is the knowledge of the physics that governs its formation and an understanding of its true nature. While glass behaves as a solid, as is experienced by all of us in our daily lives, its internal disordered structure is more reminiscent of a liquid. In an industrial society that prides itself with using scientific principles to guide the production of goods, research on the nature of glass could have profound real world consequences for industries.

With their now famous paper O'Hern et al. [14], suggested a way forward for to understand the nature of glass. In the process they also ended up trying to link two particularly intriguing questions in separate branches of physics - how glass transition happens in solid state physics, and what is the nature of granular materials in statistical physics. The gist of their idea behind linking these two areas of physics, was that one could argue that in both these systems there is a *transition* from a *flowing state* to one that was stuck or *jammed*. With the benefit of hindsight, we know that for glass formation this scenario isn't generally applicable [11] , but this landmark paper has lead to a rapidly growing branch in soft matter physics called *jamming*, that has wide-spread implications for understanding colloids, granular matter and emulsions.

Our focus is on systems like emulsions and foams in which you have one or more liquids dispersed in another fluid that acts as the dispersion medium. These can be modelled as weakly attractive collection of soft, frictionless particles as opposed to the usual model used in jamming, i.e. soft, frictionless particles. We shall assume henceforth, that the collection of particles dealt with in this document are frictionless. We shall go through the most important results from jamming for soft particles in chapter 1 . Then, we will introduce the model for weakly attractive, soft particles in chapter 2. We study the linear response obtained by perturbing the position of particles, and by introducing a shear degree of freedom for the confining box of the particles in chapter 3. The presence of attraction in weakly attractive particles and its effect on linear response is studied in chapters 4 and 5. Finally, we study the nonlinear response of finite, quasi-statically shear straining packings that have weakly attractive particles in chapter 6.

1.2. Introduction

The *collection* of particles that we were discussing in the previous section has a unifying property. They happen to be disordered, as opposed to systems like crystals, which have

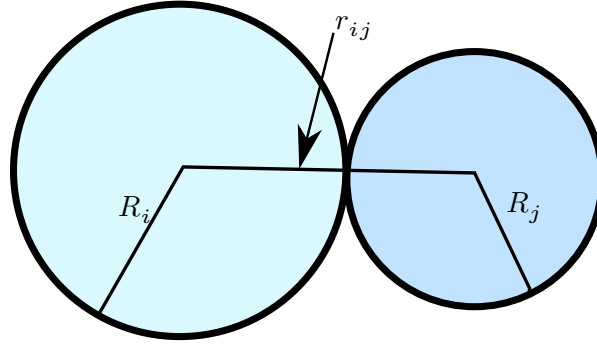


Figure 1.1: Particles i and j , with radii R_i and R_j , respectively, at a distance of r_{ij} from each other. This is representative of a contact in a packing.

structures that repeat themselves at regular intervals. We can therefore expect that disordered systems will have different properties and behaviour as compared to ordered systems [18].

In physics, we come across many systems that could be considered to be liquid because they *flow*; but under just the right conditions, they *jam* and behave as a solid. For example, it is observed that low viscosity liquids, which could be considered to be a collection of disordered molecules, undergo a transition to glass at low temperatures. More commonly, we know from experience that ketchup in a bottle sometimes requires the bottle to be shaken for it to flow freely. We are essentially applying a shear stress on the bottle in order to make ketchup flow. Clearly temperature, and the stress applied on such systems appear to affect their transition from a state in which they flow to a state in which they jam.

A new jamming phase diagram was introduced by Liu and Nagel [7], that took into consideration the temperature, applied stress and the volume fraction. At the point where $T = 0$ and zero applied shear stress, point J , the transition between jammed and unjammed is sharply defined [13], and this is the point that is studied by O'Hern et al. [14]. Point J is a critical point, since it governs the behaviour of the jammed/unjammed surface in the phase diagram [14].

1.3. Model Overview and Assumptions

In this section we'll introduce the canonical model that is used in literature for studying the jamming transition. A 2-D box, with periodic boundary conditions is filled with frictionless, spherical particles, that interact through a short-range, finite potential. There are two types of particles present in the box. They have their radii in the ratio of 1 : 1.4. They are present in a 50-50 ratio in the box; it is a bi-disperse mixture. The odd radii ratio for the particles is taken so as to avoid crystallization and thus ensure that there is disorder in the system [14]. Due to finite size effects, the number of particles matter [5]. Unless stated otherwise, the number of particles in the box is 1024. This collection of particles will be called a packing. Let us assume two particles in this box, particles i and particle j , as shown in figure 1.1. The distance between their centres is represented by r_{ij} , and the sum of their radii can be expressed by $R_{ij} = R_i + R_j$. The particles interact because of a potential that exists between them.

If we take an energy scaling factor called ϵ into consideration, and think of this potential as one that derives from Hooke's law (see figure 1.2), then the potential with which the particles

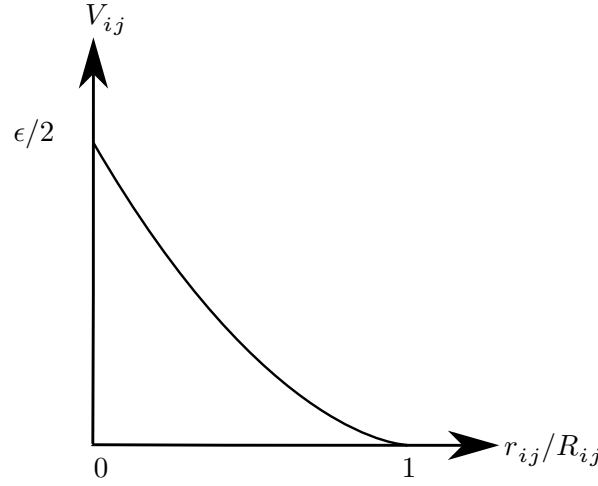


Figure 1.2: The representative illustration of the potential V_{ij} that exists between soft, repulsive particles i and j in a packing

interact can be written as:

$$V_{ij} = \begin{cases} \frac{\epsilon}{2} \left(1 - \frac{r_{ij}}{R_{ij}}\right)^2 & , \text{if } \frac{r_{ij}}{R_{ij}} \leq 1 \\ 0 & , \text{if } \frac{r_{ij}}{R_{ij}} > 1 \end{cases}$$

We are restricted to the situation in which the particles only interact if they overlap with one another. For convenience, we define a new term δ_{ij} , called the relative overlap between two particles i and j :

$$\delta_{ij} = \left(1 - \frac{r_{ij}}{R_{ij}}\right)$$

It is a dimensionless quantity, which tells us how much of an overlap exists between two particles with respect to the case when they simply touch each other.

The potential can be rewritten as:

$$V_{ij} = \begin{cases} \frac{\epsilon}{2} \delta_{ij}^2 & , \text{if } \delta_{ij} \geq 0 \\ 0 & , \text{if } \delta_{ij} < 0 \end{cases} \quad (1.1)$$

Now, to get the force of interaction between the two particles, we simply take the gradient of the potential that exists between them, and as we shall see this will give us a familiar force (see figure 1.3) that resembles the one that we get from Hooke's law:

$$F_{ij} = -\frac{dV_{ij}}{dr_{ij}} \quad (1.2)$$

$$F_{ij} = \begin{cases} \frac{\epsilon \delta_{ij}}{R_{ij}} & , \text{if } \delta_{ij} \geq 0 \\ 0 & , \text{if } \delta_{ij} < 0 \end{cases} \quad (1.3)$$

We also get the following expression for the effective spring constant that exists between the

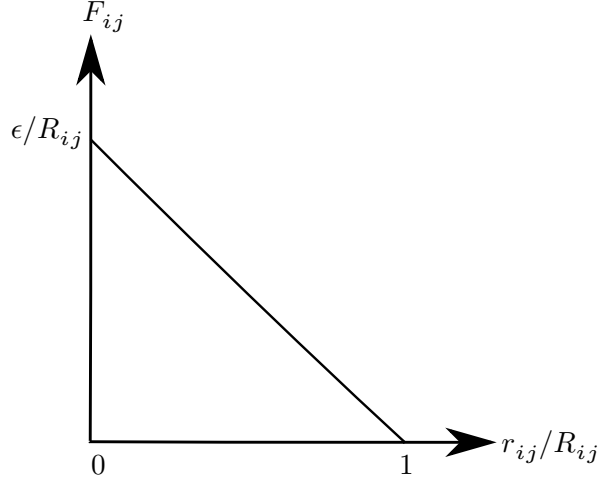


Figure 1.3: The representative illustration of the force F_{ij} between particles i and j

two particles. This is simply the gradient of the force that exists between the two particles:

$$k_{ij} = -\frac{dF_{ij}}{dr_{ij}} \quad (1.4)$$

$$= \frac{d^2V_{ij}}{dr_{ij}^2} \quad (1.5)$$

$$k_{ij} = \begin{cases} \frac{\epsilon}{R_{ij}^2} & , \text{if } \delta_{ij} \geq 0 \\ 0 & , \text{if } \delta_{ij} < 0 \end{cases} \quad (1.6)$$

Temperature fluctuations at room temperature do not overtly affect the properties of a granular system [13]. Therefore, the temperature of our systems is taken to be at the thermodynamic limit of $T = 0$ K. The particles are put at random points in the box which means that the packing is at a temperature $T = \infty$. Initially random states are sampled by looking at an ensemble of many such packings. The energy of the packings is minimized, using a conjugate gradient routine, by following the steepest gradient of the potential energy surface that represents the energy of interactions of all particles. We are *quenching* all the particles from their initial random positions to an energy minimum which would represent that they are at $T = 0$ K.

1.4. Jamming Transition

In this section, we look at how we can characterize the *jamming transition*. At the onset of this transition to being jammed, the particles would have to develop a self supporting mechanism. These particles aren't bound by anything, so the only way they can make their presence felt is by ensuring that they have a self-supported mechanism that is based on the interaction potential of the particles [18]. It is achieved by forming a network of contacts between the particles. This is shown in figure 1.4. When this happens, we would also notice that the pressure exerted by this network of particles on the environment around it, becomes non-zero. Thus a jamming transition, is necessarily a transition from a packing that has non-overlapping particles, to one that has barely touching particles. We are therefore interested in a marginally jammed packing, one where particles are just about touching each other, since it leads to interesting physics. In the next section, the bulk properties that affect the jamming transition are discussed.

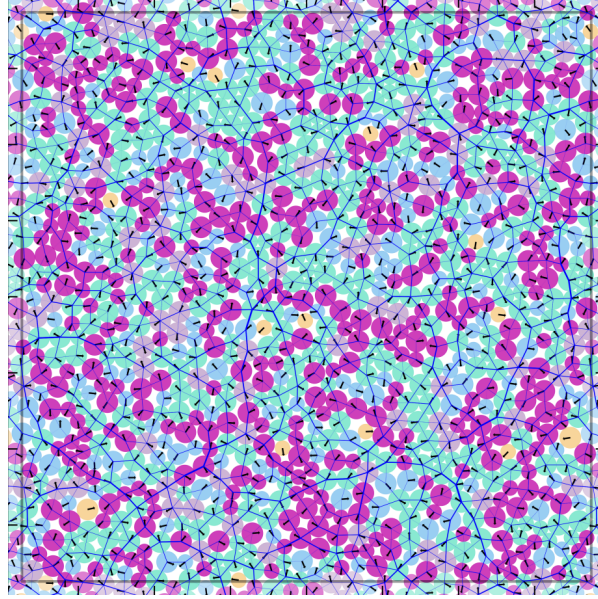


Figure 1.4: Force network (in blue) for soft, frictionless particles in a packing above the jamming point J . The thickness represents the magnitude of the repulsive force between two particles. The particles are given a certain color according to the number of contacts they have attained - light pink (6 contacts), cyan green (5 contacts), magenta (4 contacts), light blue (3 contacts), orange (≤ 2 contacts). The arrows show the displacement of the particles at a low energy mode (a soft mode). It is interesting to note the non-affine behaviour of these displacements. This is talked about in chapter 4

Contact Number

A particle will be in contact with another if it barely touches or overlaps with the other particle. In other words, the relative overlap between the particles, $\delta_{ij} \geq 0$. The contact number, which is denoted by Z is the average number of such contacts that a particle makes in a packing. So, by definition [14], it is denoted by :

$$Z = \frac{\sum_{i>j} \mathbb{1}_{ij}}{N}$$

where :

$$\mathbb{1}_{ij} = \begin{cases} 0 & , \text{if } \delta_{ij} \leq 1 \\ 1 & , \text{if } \delta_{ij} > 1 \end{cases}$$

and N denotes the total number of particles in a packing.

There is a certain number of contacts that are required to ensure that there is rigidity in the system. As an example let us take three particles that crowd around a particle. There can exist an opening for this particle to move away from the others without costing any elastic energy. This is called a *floppy mode* [18], and the particle that can move without costing elastic energy is called a *rattler* and therefore not contributing to rigidity. This simple example illustrates why the number of contacts would play a role for rigidity in a system. There can also be angular requirement for contacts which don't lead to floppy modes. If four particles crowd around one particle, but they happen to be within π radians, a floppy mode would again be possible.

So how many contacts would one need to ensure rigidity and no floppy modes ? We know that all contacts would contribute to a change in elastic energy. This number of these contacts is $ZN/2$, the factor of half ensures that we don't double count the number of contacts. The degrees of freedom for all particles in the packing is dN , where d is the number of dimensions and N is the number of particles. We wish to ensure that the terms that contribute to elastic

energy are over and above the total degrees of freedom to ensure no floppy modes [12]. This can be thought of in terms of force balance equations. There would be $ZN/2$ number of force equations, but there are dN possible variables that represent all the N particles degrees of freedom. Unless the number of force equations exceed the degrees of freedom, it will always be possible to come up with a trivial solution for the equations [18]. We wish to avoid this situation. Therefore, we can say :

$$\begin{aligned} \frac{ZN}{2} &\geq dN \\ Z &\geq 2d \end{aligned}$$

Since we are interested in frictionless, spherical particles in 2 dimensions, this number comes out to be $Z \geq 4$ [14]. In literature, when Z is equal to $2d$, it is called the isostatic contact number, or Z_{iso} [14]. This represents the case when the degrees of freedom are exactly equal to the number of contacts in a packing. Geometrically, it would mean that the particles are just about touching each other, and that they are *marginally jammed*. When particles start overlapping with each other, we find that $Z > Z_{iso}$ in a jammed packing. There may exist some particles that have $Z \leq 3$ in jammed packings. They are rattlers, as we noted previously. They do not contribute to a change in elastic energy and that their existence does not in any way impede the above analysis. They are simply removed from the entire process of calculating Z .

The rise of rigidity due to formation of contacts is essential for a packing to be in a jammed state [14]. But at the macro-scale, how would we go about measuring its effects ? The first quantity that comes to mind is to examine the stress tensor for a packing. This will give us a complete picture of the state of the packing. The derivation is shown in appendix A.

Stress Tensor

The stress tensor for a packing is written as [14] :

$$\sigma_{\alpha\beta} = \frac{1}{V} \sum_{i>j} F_{\alpha}^{ij} r_{\beta}^{ij} \quad (1.7)$$

where α and β represent the coordinate axes for the system, F_{α}^{ij} , represents the force component between i and j particle in direction α and r_{β}^{ij} represents the vector component between the particles i and j in the direction β , and V represents the volume of the box (in the 2-D case it means the area of a box, $V = L^2$, where L is the length of the box).

Pressure

With the stress tensor in hand, we can now measure many other macro-scale quantities. We start with the pressure p [14]. It is simply represented as :

$$p = \frac{\sum_{\alpha} \sigma_{\alpha\alpha}}{d} \quad (1.8)$$

$$= \frac{\sum_{\alpha} \sigma_{\alpha\alpha}}{2} \quad (1.9)$$

where σ is the stress tensor, and d represents the dimension of the packing. In our 2-D case, this is $d = 2$. The rise of rigidity in our system, would require that there is a jump in the pressure that the jammed system exerts, from zero to a non-zero value.

Packing Fraction

The volume of space occupied by the particles in a packing as a fraction of the total volume is defined as the packing fraction for a packing. The packing fraction is represented by ϕ in literature [14], and can be written as :

$$\begin{aligned}\phi &= \frac{\sum_i V_i}{L^d} \\ &= \frac{\sum_i \pi R_i^2}{L^2}\end{aligned}$$

where i is the i^{th} particle, V_i is the volume of the i^{th} particle which in a 2-D case means its area, d is the dimension of the packing, L^d is the volume of the box, which in a 2-D case is L^2 .

For each packing there is a critical packing fraction at which the packing is jammed, and it is denoted by ϕ_c . At this critical volume fraction, the average number of contacts become Z_{iso} , and the contacts ensure that there is rigidity, and that the particles now exert a non-zero pressure. We note, however that the critical volume fraction can be slightly different for every packing, since for every packing at the jamming transition, we sample one of the many available energy minimums that would lead to a marginally jammed packing. It isn't therefore uniquely representative of the jammed state, unlike pressure, which will make a sudden jump from zero to non-zero at the jamming transition for every marginally jammed packing.

Bulk Modulus

Now that we have pressure and volume fraction defined, we can formalize the definition of the bulk modulus for our system. Bulk modulus is simply the measure of how resistant is the packing to a change in volume or a change in density. The volume fraction ϕ is a good measure for a change in volume for our system. This can be written as [14] :

$$B = -\phi \frac{dp}{d\phi}$$

As the distance from ϕ_c increases towards the jammed state, the bulk modulus of the packing increases as well.

Shear Modulus

The shear modulus measures the resistance of a packing when we apply a shear strain at its boundaries. It can be noted down as :

$$G = \frac{d\sigma_{xy}}{d\gamma}$$

where σ_{xy} is the xy component of the stress tensor, and γ is the shear strain in the x direction, and with a gradient in the y direction.

1.5. Scaling Arguments

As noted earlier, the jamming point is a critical point. Scaling arguments provide us with critical exponents that can describe the behaviour of marginally jammed systems at this critical point. We shall discuss the scaling of the quantities discussed previously as a distance to jamming. As we have already noted, distance to jamming can either be measured by pressure, i.e. when the pressure of a packing becomes non-zero it can be considered to be jammed. Or, we can take $\phi - \phi_c$ as our criterion for measuring the distance to jamming, with the understanding that ϕ_c can be different for individual packings.

Pressure

It has been found that in the case of 2-D bi-disperse mixtures [14] :

$$p \sim \phi - \phi_c \quad (1.10)$$

$$p \sim \Delta\phi \quad (1.11)$$

This can be understood from the fact that the pressure is proportional to force which we get from the second derivative of the potential. Hence $p \sim F$. In addition, we also know that the quantity $\phi - \phi_c$ represents the overlap between particles (remember that ϕ_c is the isostatic point), and since the force is directly proportional to the overlap, we can say that $\phi - \phi_c \sim F$.

Contact Number

At the jamming transition, it has been found to have the following scaling [14, 18] :

$$\begin{aligned} Z - Z_c &\sim (\phi - \phi_c)^{1/2} \\ \Delta Z &\sim (\Delta\phi)^{1/2} \end{aligned}$$

The contact number jumps discontinuously to the isostatic value at the critical packing fraction. It has been found that this scaling is independent of the polydispersity of the system [14]. We assume a mono-disperse system, for simplification, with particles of radius 1, for which it has been found that the radial distribution function [16, 18] :

$$g(r) \sim \frac{1}{\sqrt{r-1}}$$

diverges at the jamming transition, i.e. at $r = 1$, since the radii of particles is 1. This may be understood as particles just about touching each other at the isostatic point, and therefore crowding about and producing a delta function for $g(r)$ at $r \rightarrow 1$. The area under the curve of $g(r)$ will correspond to the number of contacts that are present in the packing. At the isostatic point this would be then equal to $Nd/2$ contacts. Now, if we try to compress this packing, we go from a typical particle overlap of 0 at the isostatic point to some value which we call as δ . The marginal change in the typical contact number can be expressed in terms of the radial distribution function [16, 18] :

$$\begin{aligned} \Delta Z &\sim \int_1^{1+\delta} \frac{1}{\sqrt{r-1}} dr \\ \Delta Z &\sim \sqrt{\delta} \end{aligned}$$

This gives us a scaling for the overlap length scale from the previous discussion on Z . This is written as $\delta \sim \Delta\phi$.

Bulk Modulus

As has been discussed above the bulk modulus is found from the derivative of the pressure with respect to the packing fraction, and it has been shown that the bulk modulus changes in an affine manner as a function of the distance to the jamming transition for 2-D, bi-disperse packings [14] :

$$B \sim (\phi - \phi_c)^0$$

Shear Modulus

It has been shown that the shear modulus does not follow the scaling that we would predict from an affine assumption [14]. We shall discuss the non affine nature of disordered systems in detail in chapter 4. The shear modulus of marginally jammed packings follows the following scaling :

$$G \sim (\phi - \phi_c)^{1/2}$$

Note that the elastic moduli and pressure shown here are scale as power laws with the distance to the jamming point ϕ_c , which is why it has been described as a critical point [14]. The discontinuous jump in the contact number at the jamming point and the continuous increase in pressure from zero at the jamming point J means that the jamming transition has characteristics of both first and second order transitions [8, 14]. We now have the basic quantities that describe and govern behaviour of packings at the jamming transition. They will help us in understanding the results of this report. The physical quantities discussed here have units that derive from the effective stiffness of a contact, which for our potential scales with ϵ , the energy scaling factor [14, 18] if we consider the units for the radii as one. In the next chapter, we shall discuss the model for weakly attractive particles.

2

Weakly Attractive Particles

2.1. A Model for Particles with Attraction

As we discussed in section 1.1, the model for this work is for systems like emulsions and foams. We shall extend the model for soft, frictionless particles that we had used in chapter 1, so as to hold for our system of interest. We use emulsions as an example to understand the origin of the weakly attractive forces between particles that exist in foams and emulsions. Emulsions are formed when one liquid is dispersed in another liquid. For example, let us take oil droplets that are dispersed in water. As they are immiscible liquids, this emulsion also needs a surfactant that keeps the liquids mixed with each other. Surfactants usually have long carbon chains as *tails* that are *hydrophobic*, and have a *head* that is *hydrophilic*. The hydrophobic part dissolves in oil, and the hydrophilic part dissolves in water. Surfactants increase the stability of emulsions, by effectively decreasing the surface tension between the constituent dispersed liquids, and preventing droplet coalescence. This is shown in figure 2.1.

We can think of the dispersed oil droplets in water as soft spheres. In addition, we now need to model the consequence of adding the surfactant. The addition of the surfactant would mean that the oil droplets have a weakly attractive force around them, due to *depletion interaction*. We extend this idea of a weakly attractive force around soft spheres to the model we had used previously for repulsive soft spheres. We can do this by imagining a thin shell around a soft sphere. This shell is the volume within which the attractive force exists.

Let us take a pair of particles i and j with radii R_i and R_j respectively, as in figure 2.2. The distance between their centres, is represented by r_{ij} . However, there is an imaginary shell around both these particles. For particle i , this shell extends from R_i to $R_i(1 + 2a)$, where a

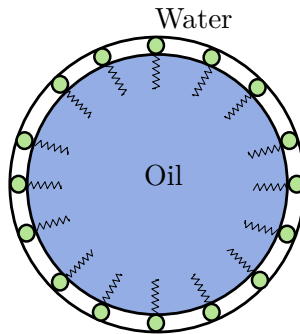


Figure 2.1: An oil droplet in water as part of an emulsion is shown here. The hydrophilic head of the surfactant is dissolved in the water, and the hydrophobic tail is dissolved in the oil.

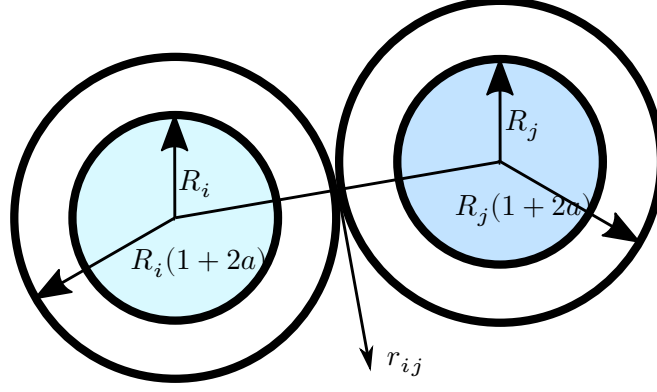


Figure 2.2: Weakly attractive shells surround particles i and j . In this picture, the shells of the particles, and not the particles themselves, are just about touching each other.

is a tunable attraction parameter, that represents the attraction strength between the spheres. Similarly, such a shell also exists for particle j . This is shown in figure 2.2.

The interaction potential, V_{ij} between this pair of particle is of the following form [6, 9, 20]:

$$V_{ij} = \begin{cases} \frac{\epsilon}{2} \left[\left(1 - \frac{r_{ij}}{R_{ij}} \right)^2 - 2a^2 \right] & , \text{ if } \frac{r_{ij}}{R_{ij}} < 1 + a \\ -\frac{\epsilon}{2} \left[1 + 2a - \frac{r_{ij}}{R_{ij}} \right]^2 & , \text{ if } 1 + a < \frac{r_{ij}}{R_{ij}} < 1 + 2a \\ 0 & , \text{ if } \frac{r_{ij}}{R_{ij}} > 1 + 2a \end{cases} \quad (2.1)$$

This is reminiscent of the repulsive potential with the addition of an attractive tail added to it. We also note that we can define a relative overlap in the case of attraction, which we can call as δ_{ij}^{attr} :

$$\delta_{ij}^{attr} = 1 + 2a - \frac{r_{ij}}{R_{ij}}$$

The potential that we consider gives us a force F_{ij} between particles i and j , and it can be written as :

$$F_{ij} = \begin{cases} \frac{\epsilon}{R_{ij}} \left[1 - \frac{r_{ij}}{R_{ij}} \right] & , \text{ if } \frac{r_{ij}}{R_{ij}} < 1 + a \\ -\frac{\epsilon}{R_{ij}} \left[1 + 2a - \frac{r_{ij}}{R_{ij}} \right] & , \text{ if } 1 + a < \frac{r_{ij}}{R_{ij}} < 1 + 2a \\ 0 & , \text{ if } \frac{r_{ij}}{R_{ij}} > 1 + 2a \end{cases} \quad (2.2)$$

In addition, the stiffness between the particles, k_{ij} , can be written as :

$$k_{ij} = \begin{cases} \frac{\epsilon}{R_{ij}^2} & , \text{ if } \frac{r_{ij}}{R_{ij}} < 1 + a \\ -\frac{\epsilon}{R_{ij}^2} & , \text{ if } 1 + a < \frac{r_{ij}}{R_{ij}} < 1 + 2a \\ 0 & , \text{ if } \frac{r_{ij}}{R_{ij}} > 1 + 2a \end{cases} \quad (2.3)$$

We note that as $a \rightarrow 0$, the model described above, would be the same as the model for repulsive particles as we had discussed in chapter 1. The model for weakly attractive, soft and

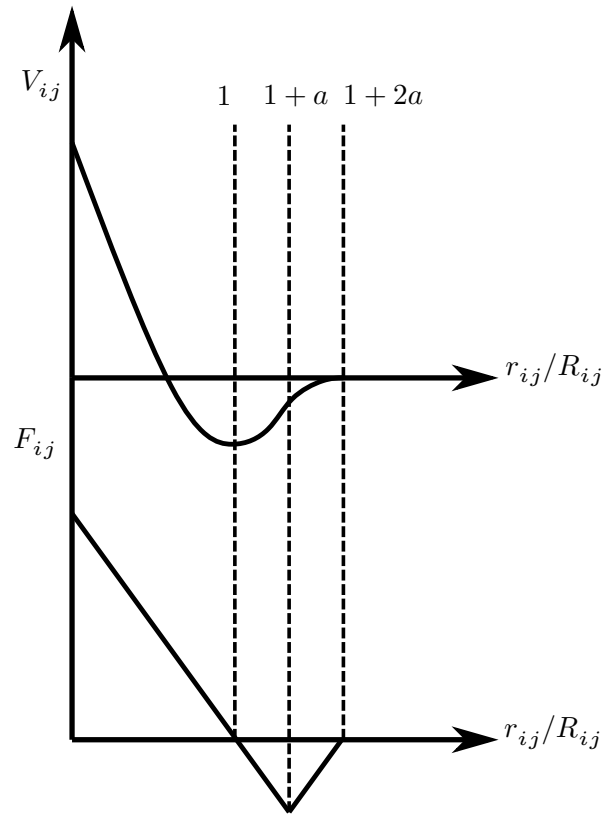


Figure 2.3: Potential, V_{ij} and force, F_{ij} between weakly attractive particles i and j

frictionless particles is used in chapter 4 and chapter 6. In the next chapter, we look at the linear response of soft, frictionless particles by perturbing the initial positions of the particles by a small amount, and also by adding a shear strain degree of freedom for the box within which the particles are confined.

3

Linear Response to Perturbations

What happens when particles in a packing are perturbed from their initial positions by a small amount ? To investigate this, let us take two soft, frictionless particles i and j just about touching each other and assume that they behave as a spring [2, 19]. For a packing, this would mean replacing contacts with springs [19]. This assumption inherently also means that no new contacts in the packing are made nor are the existing ones broken. Our assumed spring would then have been at a relaxed length of l_0 , which would be the sum of the radii of the two particles. This spring, is known to have a spring constant k . They are displaced by $\mathbf{u}_1$ and $\mathbf{u}_2$ respectively, as shown in 3.1. We note down the important parameters of this initial configuration of the two particles. Here E_i is the potential energy that exists when two particles are in contact, and F_0^{ij} is the force that exists initially, before the particles are displaced.

$$E_i = \frac{k}{2}(l - l_0)^2 \quad (3.1)$$

$$F_0^{ij} = -k(l - l_0) \quad (3.2)$$

Next, we move to a frame where the first particle is fixed. So the displacement of the second particle, in the new frame, becomes $\mathbf{u}_{21} = \mathbf{u}_2 - \mathbf{u}_1$. We decompose u_{21} , parallelly and orthogonally to line that connects the initial centres of the particles. They are called $\mathbf{u}_{21\parallel}$ and $\mathbf{u}_{21\perp}$, respectively. This is shown in figure 3.2. The new length of the spring, after the

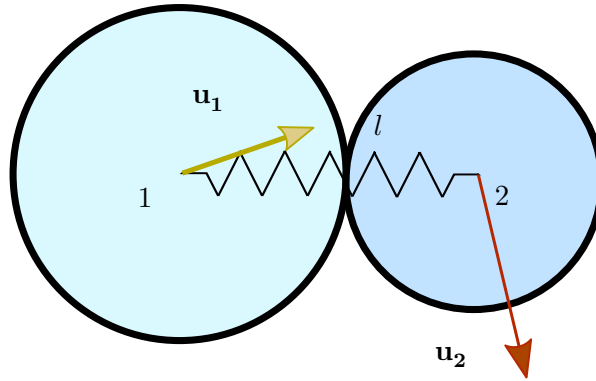


Figure 3.1: The particles in this illustration are displaced by a certain amount $\mathbf{u}_1$ and $\mathbf{u}_2$ from their initial configuration

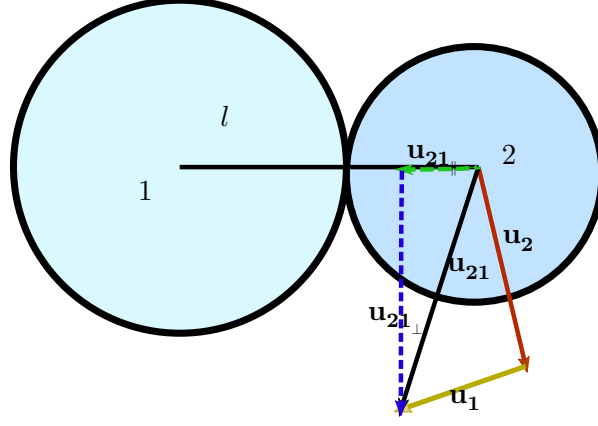


Figure 3.2: We assume that particle 1 is our frame of reference. In this frame, the relative displacement of particle 2 is decomposed along the line that connects the centre of the two particles initially, and thus find $\mathbf{u}_{\parallel}$ and $\mathbf{u}_{\perp}$.

displacements, becomes l' . This can be expressed in the following way [18] :

$$l' = \sqrt{(l + u_{21\parallel})^2 + u_{21\perp}^2} \quad (3.3)$$

$$l' = l \sqrt{\left(1 + \frac{u_{21\parallel}}{l}\right)^2 + \left(\frac{u_{21\perp}}{l}\right)^2} \quad (3.4)$$

When this expression is written down as a Taylor expansion, and terms of order 3 and higher are neglected, we see that :

$$l' = l \left[1 + \frac{u_{21\parallel}}{l} + \frac{1}{2} \left(\frac{u_{21\perp}}{l} \right)^2 \right] \quad (3.5)$$

The new energy E_f , of our assumed spring is now :

$$E_f = \frac{1}{2} k (l' - l_0)^2 \quad (3.6)$$

The change in energy of the spring from its initial configuration and its final configuration is :

$$\delta E = E_f - E_i \quad (3.7)$$

$$= \frac{1}{2} k (l' - l_0)^2 - \frac{k}{2} (l - l_0)^2 \quad (3.8)$$

By only keeping the quadratic terms, we end up with :

$$\delta E = \frac{k u_{21\parallel}^2}{2} - F_0^{ij} \left(u_{21\parallel} + \frac{u_{21\perp}^2}{2l} \right) \quad (3.9)$$

This needs to be extended for a many particle system, the change in energy of a many particle system ΔU can be written as [18, 19] :

$$\Delta U = \sum_{\langle i,j \rangle} \delta E_{ij} = \sum_{\langle i,j \rangle} \frac{k_{ij}}{2} \left[u_{ij\parallel}^2 - F_0^{ij} \frac{u_{ij\perp}^2}{k_{ij} l_{ij}} \right] \quad (3.10)$$

The sub-indices, in the terms above, reflect the pair of particles that are in contact in the many particles system. To get force equations for our system, we use Lagrangian mechanics. We define,

$$L = T - U \quad (3.11)$$

where L , T , U are the Lagrangian, the kinetic energy and the potential energy respectively. In our system the degree of freedom are $\mathbf{q} = \{u_1^x, u_2^x, u_3^x \dots, u_1^x, u_2^y, u_3^y \dots\}$, where u_i^α is the i^{th} particle displacement in the α direction. We assume that the particles can only move in x and y directions. Since the temperature is taken to be zero, the kinetic energy of our system is zero and thus $L = -\Delta U$. When we write the Euler-Lagrange equation for this system, we need to take into account the fact that it is a quasi-static system. Therefore,

$$-\frac{\partial L}{\partial q_i^\alpha} = F_i^\alpha \quad (3.12)$$

$$F_i^\alpha = \frac{\partial \Delta U}{\partial q_i^\alpha} \quad (3.13)$$

In the expression for ΔU in equation 3.10, we see that relative displacements between particles in contact are used. We can always express them in terms of absolute displacements, which is shown in section 3.1. Now, we note that when we do this, ΔU will be a quadratic expression in terms of absolute displacements.

$$\Delta U = \frac{1}{2} \sum_{\langle i, j \rangle} \sum_{\langle \alpha, \beta \rangle} K_{ij}^{\alpha\beta} q_i^\alpha q_j^\beta \quad (3.14)$$

$$\Delta U = \frac{\mathbf{q}^T \mathbf{K} \mathbf{q}}{2} \quad (3.15)$$

In solid mechanics' terminology, $\mathbf{K}$ can be seen as the *stiffness matrix*. Physicists call it the *dynamical matrix* [14, 18]. When we simplify the force relations, after substituting the above representation of the potential energy, we see that $\mathbf{K}$ is essentially a Hessian matrix of the form given below:

$$K_{ij}^{\alpha\beta} = \frac{\partial^2 U}{\partial q_i^\alpha \partial q_j^\beta} \quad (3.16)$$

This makes sense, since the packing is at a one of the many available energy minimums. If we were to Taylor expand the perturbed energy of the packing, the leading contribution to the change in energy - when we assume that the initial energy of the packing is zero - would be from the Hessian term of the expansion, as the Jacobian term would be zero (i.e. $\nabla U = 0$) [14, 17].

3.1. Relative to Absolute Coordinates

If the initial position of the particles in contact are expressed as $\mathbf{r}_i, \mathbf{r}_j$, we can now find the angle that the displacement vector $\mathbf{u}_{ij} = \mathbf{u}_i - \mathbf{u}_j$ makes with the vector $\mathbf{r}_{ij} = \mathbf{r}_i - \mathbf{r}_j$ that initially connects the centres of the particles.

$$\cos \theta = \frac{\mathbf{r}_{ij} \cdot \mathbf{u}_{ij}}{|\mathbf{r}_{ij}| |\mathbf{u}_{ij}|} \quad (3.17)$$

$$= \frac{U_{ij}^x X_{ij} + U_{ij}^y Y_{ij}}{r_{ij} u_{ij}} \quad (3.18)$$

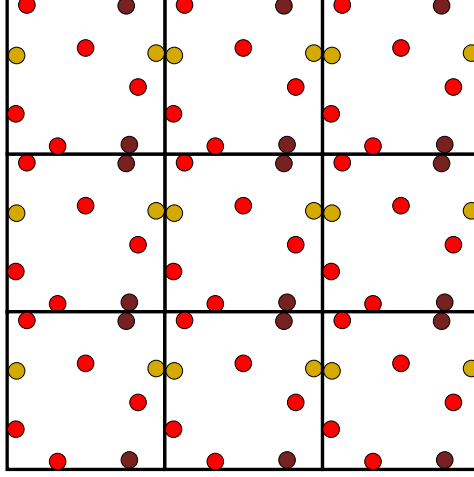


Figure 3.3: Periodic boundary conditions for the packing. Copies of the packing in the center of the illustration are placed around the four sides of the box. This would introduce differences in the way that the coordinates of particles in contact, especially near the boundaries, are calculated.

where the symbols mean the following :

$$U_{ij}^x = u_i^x - u_j^x \quad (3.19)$$

$$U_{ij}^y = u_i^y - u_j^y \quad (3.20)$$

$$X_{ij} = x_i - x_j \quad (3.21)$$

$$Y_{ij} = y_i - y_j \quad (3.22)$$

$$r_{ij} = |\mathbf{r}_{ij}| \quad (3.23)$$

$$u_{ij} = |\mathbf{u}_{ij}| \quad (3.24)$$

In the first term of 3.10, we have the term $u_{ij\parallel}^2$. We can write this simply as :

$$u_{ij\parallel}^2 = u_{ij}^2 \cos^2 \theta \quad (3.25)$$

$$= \left(\frac{U_{ij}^x X_{ij} + U_{ij}^y Y_{ij}}{r_{ij}} \right)^2 \quad (3.26)$$

Similarly, we can find that :

$$u_{ij\perp}^2 = \left(u_{ij}^2 - u_{ij\parallel}^2 \right) \quad (3.27)$$

$$= u_{ij}^2 - \left(\frac{U_{ij}^x X_{ij} + U_{ij}^y Y_{ij}}{r_{ij}} \right)^2 \quad (3.28)$$

In the following sections, we will discuss briefly the implications of introducing boundary conditions to the displacements of particles and the subsequent changes in expressions for the dynamical matrix or as we'll call it henceforth, the Hessian.

3.2. Periodic Boundary Conditions

A periodic boundary condition is used when we want to remove the effects of having a wall at the four faces of the box that makes our packing. It works by copying the packing, and placing it around the original packing, as shown in Fig. 3.3 [14]. A contact between two repulsive

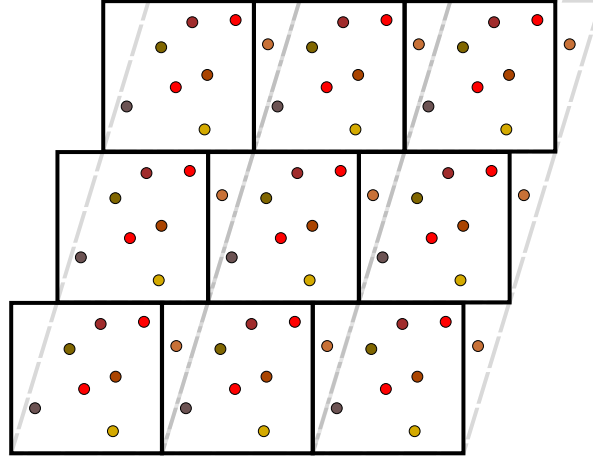


Figure 3.4: Lees-Edwards boundary conditions for a sheared packing. Copies of the sheared central packing are placed along the boundaries, but they are displaced by the amount of the shear strain that acts on the original packing.

particles i and j exists if the distance between their centres r_{ij} is :

$$r_{ij} \leq (R_i + R_j) \quad (3.29)$$

and for the case of particles with weakly attractive interaction, this becomes :

$$r_{ij} \leq (R_i + R_j)(1 + 2a) \quad (3.30)$$

In addition, we forgo those contacts that are made by rattlers. Let's assume the origin of the coordinate system lies at the center of the box. We want to find out all the contacts in the packing, taking into consideration the periodic boundary conditions. We do this in two steps :

- Check if the particles are in contact in the packing.
- Due to the periodic boundary conditions, we then need to check for contacts at the boundaries, simply by copying the packing, and placing it at the boundaries and checking for contacts.

Periodic boundary conditions will have an effect on the coordinates of the particles in contact near the boundary. They will have to be appropriately changed by adding (or subtracting) the length of the box L from the x or y coordinate of the particles depending on their positions with respect to the assumed origin. We assume the origin to be at the center of the packing, and we call it O . This is shown in 3.5. The new coordinates x' and y' , relative to origin O , can be written as :

$$\begin{aligned} x' &= x \pm L \\ y' &= y \pm L \end{aligned}$$

3.3. Lees - Edwards Boundary Conditions

In this section, we shall look at how this response changes when a box degree of freedom, namely shear strain, is introduced. Let's assume that the y box face is strained in the x direction by a vanishingly small amount of strain γ . Assume the origin of the coordinate system lies at the center of the box, as we did before in the periodic boundary conditions. This is shown in figure 3.5 In addition to fulfilling the periodic boundary condition, the particles now also have to full

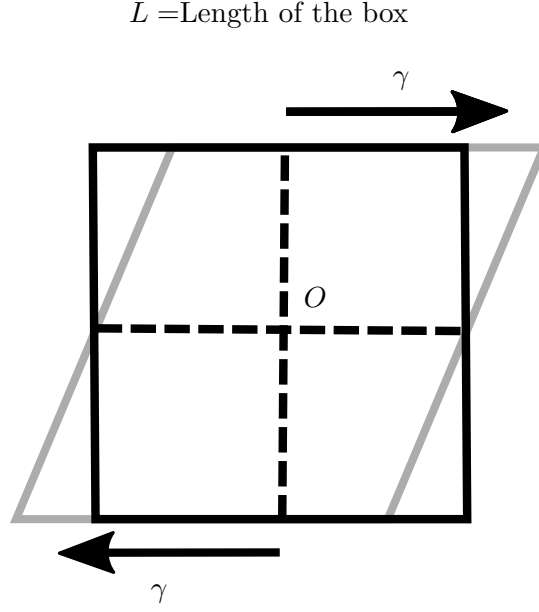


Figure 3.5: Shear Strained Packing

fill the sheared boundary condition, commonly called the Lees-Edwards boundary condition [3]. This is shown in 3.4. The y coordinates of the particles at the boundary of the packing remain the same, but their x coordinate changes to the new coordinate x' in the following way :

$$x' = x \pm \gamma L$$

depending on whether the particle is above or below the origin O , as shown in 3.5.

3.4. Changes to the Hessian

The shear strain to the box now adds another degree of freedom to $\mathbf{q}$, and therefore changes the coordinates of the particles, as we have shown in the above section. The degrees of freedom now becomes the following :

$$\mathbf{q} = \{u_1^x, u_2^x, u_3^x \dots, u_1^y, u_2^y, u_3^y \dots, \gamma\} \quad (3.31)$$

where u_i^α is the i^{th} particle displacements in the α direction, and γ is the shear strain imposed on the packing. If there are N particles, then the Hessian matrix $\mathbf{K}$ is now of the size $(2N + 1) \times (2N + 1)$. This is because of the addition of the following terms for each contact :

$$K_{ij}^{\alpha\gamma} = \frac{\partial^2 U}{\partial q_i^\alpha \partial \gamma} \quad (3.32)$$

$$K_{ij}^{\beta\gamma} = \frac{\partial^2 U}{\partial q_i^\beta \partial \gamma} \quad (3.33)$$

$$K_{ij}^{\gamma\gamma} = \frac{\partial^2 U}{\partial^2 \gamma} \quad (3.34)$$

The derived terms for the Hessian matrix are shown in appendix D. In this chapter, we have derived the linear response of packings by assuming that at every contact there exists a relaxed spring, and then perturbing the springs by a small amount, in addition to incorporating a vanishingly small shear strain degree of freedom that acts on the y boundaries of the packings. In the next chapter, we shall look at how to connect the bulk quantities measured from the linear response of the packings, to quantities related to particles that make up the packings.

4

Results : Linear Response

We create 200 packings for a range of pressures, $p = 10^{-6}, \dots, 10^{-2}$ and attraction strength, $a = 0, \dots, 10^{-2}$. Both of these quantities have been chosen to have a step size of one decade. The packings are created using the FCG algorithm (see appendix B), and the initial packing fraction from which we begin making the packings is $\phi = 0.9$. We use the absolute expressions for the potential, force and stiffness as shown in appendix C. This would change the units of the quantities that we measure in this chapter. As we had discussed in chapter 1, the physical quantities scale with the units of the effective stiffness constant (see the expressions for stiffness of a contact in equations C.4.3 and C.3.3). They would now scale solely with the energy scaling factor ϵ . It is done for convenience.

4.1. Strain Test

The energy of a shear strained packing (in the limit of $\gamma \rightarrow 0$) in terms of the shear modulus can be written as :

$$U = U_0 + \frac{1}{2}GV^2\gamma^2 \quad (4.1)$$

where U_0 is the initial energy in the packing, V is the volume of the packing (since we are dealing with 2-D packings, this is just L^2), G is the shear modulus, and γ is the shear strain. The second term can be found by assuming that the packing is deformed by a shear strain of γ assuming that it has a shear modulus of G . It goes without saying that in the linear response of a vanishingly small shear strain, we have assumed a linear elastic constitutive relationship between shear stress and shear strain. This has been measured by Wyart et al. [19] for rigidly stiff random networks made up of springs, and verified by Ellenbroek et al. [3] for systems like ours where we have athermal particles at an energy minimum for whom contacts have been replaced by springs. As the shear modulus is an unknown, we can use the Hessian matrix, derived in chapter 3, to calculate it. This is done by a *strain test*. We know that :

$$\mathbf{F} = \mathbf{K}\mathbf{q} \quad (4.2)$$

where $\mathbf{F}$ is the force vector, $\mathbf{K}$ is the dynamical matrix and $\mathbf{q}$ is the displacement vector. This can be written in an expanded form for N particles as :

$$\begin{bmatrix} F_1^x \\ F_1^y \\ \vdots \\ F_N^x \\ F_N^y \\ \sigma V \end{bmatrix} = \begin{bmatrix} K_{1,1} & K_{1,2} & \dots & K_{1,2N-1} & K_{1,2N} & K_{1,2N+1} \\ K_{2,1} & K_{2,2} & \dots & K_{2,2N-1} & K_{2,2N} & K_{2,2N+1} \\ \vdots & \vdots & \ddots & \vdots & \vdots & \vdots \\ K_{2N-1,1} & K_{2N-1,2} & \dots & K_{2N-1,2N-1} & K_{2N-1,2N} & K_{2N-1,2N+1} \\ K_{2N,1} & K_{2N,2} & \dots & K_{2N,2N-1} & K_{2N,2N} & K_{2N,2N+1} \\ K_{2N+1,1} & K_{2N+1,2} & \dots & K_{2N+1,2N-1} & K_{2N+1,2N} & K_{2N+1,2N+1} \end{bmatrix} \begin{bmatrix} u_1^x \\ u_1^y \\ \vdots \\ u_N^x \\ u_N^y \\ \gamma \end{bmatrix}$$

Note that we have assumed that the shear strain that we have imposed on the packing gives rise to a shear stress σ and therefore a force F_γ acts on the boundaries of the packing, which is then put in the appropriate place in the force vector $\mathbf{F}$ as $F_\gamma = \sigma V$; where V is the volume of the packing (in 2-D this is L^2). The entries of the matrix $K_{\alpha,\beta}$ are derived from the expressions given in appendix D for every contact $\langle i, j \rangle$, and put in the correct position in $\mathbf{K}$. In the first step of the strain test, we take all of the particle displacements to be zero (i.e. $\{u_1^x, u_1^y, \dots, u_N^x, u_N^y\} = \mathbf{0}$). The strain value is taken to be 1 (i.e. $\gamma = 1$), purely for convenience. This value doesn't affect the quantities that would be measured in this chapter. Next we solve the system of equations above for the forces (i.e. $\{F_1^x, F_1^y, \dots, F_N^x, F_N^y\}$). We use these forces to calculate the displacements using the smaller Hessian matrix $\mathbf{K}'$, in the following way :

$$\begin{bmatrix} F_1^x \\ F_1^y \\ \vdots \\ F_N^x \\ F_N^y \end{bmatrix} = \begin{bmatrix} K_{1,1} & K_{1,2} & \dots & K_{1,2N-1} & K_{1,2N} \\ K_{2,1} & K_{2,2} & \dots & K_{2,2N-1} & K_{2,2N} \\ \vdots & \vdots & \ddots & \vdots & \vdots \\ K_{2N-1,1} & K_{2N-1,2} & \dots & K_{2N-1,2N-1} & K_{2N-1,2N} \\ K_{2N,1} & K_{2N,2} & \dots & K_{2N,2N-1} & K_{2N,2N} \end{bmatrix} \begin{bmatrix} u_1^x \\ u_1^y \\ \vdots \\ u_N^x \\ u_N^y \end{bmatrix}$$

$$\mathbf{F}' = \mathbf{K}' \mathbf{q}'$$

$$\mathbf{q}' = (\mathbf{K}')^{-1} \mathbf{F}'$$

What we are essentially doing in the above test, is to first assume that there is a linear response to a shear strained packing. We calculate the forces that would act on the particles for a vanishingly small strain on the box. These calculated forces are then used to find the response of the particles in terms of displacements. These displacements can also be expressed in terms of relative displacements (i.e. $u_\parallel$ and $u_\perp$) as shown in chapter 3.

We can also find the shear modulus that results from the straining of the box and the response of the particles from the following:

$$\Delta U = \frac{GV^2\gamma^2}{2} \quad (4.3)$$

$$= \frac{\mathbf{q}^T \mathbf{K} \mathbf{q}}{2} \quad (4.4)$$

$$G = \frac{\mathbf{q}^T \mathbf{K} \mathbf{q}}{V^2\gamma^2} \quad (4.5)$$

The packings generated are put through the *strain test*. In the following sections, we calculate bulk quantities for a packing due to a linear response to shear strain and connect them to quantities that are tied to individual particles and contacts in the packing.

4.2. Shear Modulus

We know from chapter 1 that for repulsive particles :

$$G \sim (\phi - \phi_c)^{1/2} \quad (4.6)$$

$$p \sim (\phi - \phi_c) \quad (4.7)$$

For our case where we make packings using pressure, we can use the following scaling :

$$G \sim p^{1/2} \quad (4.8)$$

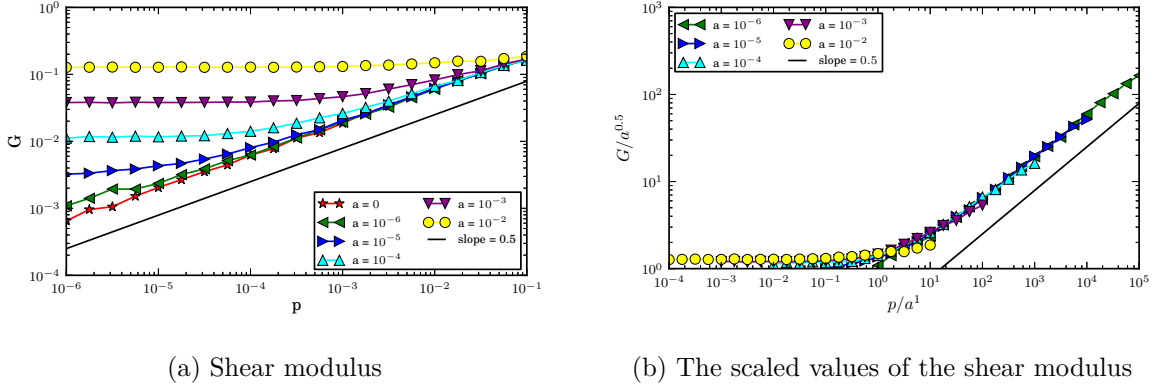


Figure 4.1: The shear modulus is obtained from assuming a linear response to a shear strained packing. This is done for a range of attraction strengths a and pressure p . The scaled value of the shear modulus is also shown.

Critical Scaling Analysis

Let's assume ansatz that :

$$p^* \sim a^\nu \quad (4.9)$$

$$G^* \sim a^\lambda \quad (4.10)$$

Its a reasonable assumption to have the shear modulus be a function of pressure and attraction as those are our varying parameters in our numerical experiments.

$$G = G(p, a) \quad (4.11)$$

$$\frac{G}{G^*} = \mathcal{G}\left(\frac{p}{p^*}\right) \quad (4.12)$$

$$\frac{G}{a^\lambda} = \mathcal{G}\left(\frac{p}{a^\nu}\right) \quad (4.13)$$

$$\frac{G}{a^\lambda} \sim \left(\frac{p}{a^\nu}\right)^\beta \quad (4.14)$$

In order to satisfy the scaling of our data, we can write the following for the shear modulus of the packing :

$$G \sim \begin{cases} a^\lambda & \text{if, } \frac{p}{a^\nu} \ll 1 \\ a^{\lambda-\beta\nu} p^\beta & \text{if, } \frac{p}{a^\nu} \gg 1 \end{cases} \quad (4.15)$$

If this relation holds true for all attraction values, including $a = 0$, then the following is true for the repulsive case :

$$\beta = \frac{1}{2} \quad (4.16)$$

$$\lambda - \nu\beta = 0 \quad (4.17)$$

$$\lambda = \frac{1}{2}\nu \quad (4.18)$$

We take the mean of the values of the shear modulus obtained from the strain test for various pressures and attraction strength. This is shown in figure 4.1a. For the case of packings with repulsive particles, we see an agreement between the theoretical scaling of G with p . In the presence of attraction, we see that with decreasing pressure there is a plateau in the value of

G . The plateauing begins at a larger pressure for a larger value of attraction. From our scaled data for the shear modulus in figure 4.1b, we find that $\nu = 1$, and $\lambda = 1/2$, and these also satisfy the relationship between them, as required by the theoretical scaling.

$$\frac{G}{a^{1/2}} \sim \left(\frac{p}{a^1}\right)^{1/2} \quad (4.19)$$

We get the following behaviour for the shear modulus from the scaled data :

$$G \sim \begin{cases} a^{1/2} & \text{if, } \frac{p}{a} \ll 1 \\ p^{1/2} & \text{if, } \frac{p}{a} \gg 1 \end{cases} \quad (4.20)$$

4.3. Relative Displacements

In addition we can also use the above relation to know how the $u_{\parallel}$ and $u_{\perp}$ scale with respect to pressure for repulsive systems. From equations 3.10 and 4.3, we can write [3, 4] :

$$G\gamma^2 \sim ku_{\parallel}^2 \quad (4.21)$$

$$G\gamma^2 \sim Fu_{\perp}^2 \quad (4.22)$$

where k is the stiffness constant of a contact, $u_{\parallel}$ is the relative displacement along the line that joins the center of the particles in contact, $u_{\perp}$ is the relative displacement perpendicular to the line of contact, and F is the initial force between the particles in contact, and G is the shear modulus. In our case, $\gamma = 1$, and therefore drops out of the equations above. The stiffness constant, k is an $\mathcal{O}(1)$ term compared to the magnitude of G and $u_{\parallel}^2$. The physical meaning of this scaling comes from the change in energy of a contact, that has been linked to the scaling of the shear modulus, a bulk quantity of the packing. Using the scaling of G and p , we find that for soft particles, the following scaling holds as we reduce the distance to the jamming transition [3] :

$$G \sim u_{\parallel}^2 \quad (4.23)$$

$$u_{\parallel}^2 \sim p^{1/2} \quad (4.24)$$

$$u_{\parallel} \sim p^{1/4} \quad (4.25)$$

We know that the Force scales proportionally to pressure, therefore using the previous scaling :

$$G \sim Fu_{\perp}^2 \quad (4.26)$$

$$G \sim pu_{\perp}^2 \quad (4.27)$$

$$u_{\perp}^2 \sim p^{-1/2} \quad (4.28)$$

$$u_{\perp} \sim p^{-1/4} \quad (4.29)$$

Critical Scaling Analysis

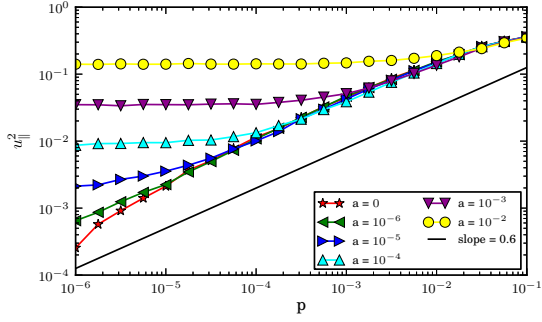
Using the same method as shown in the previous section, we can write the following :

$$u_{\parallel}^2 \sim a^{\lambda' - \beta' \nu'} p^{\beta'} \quad (4.30)$$

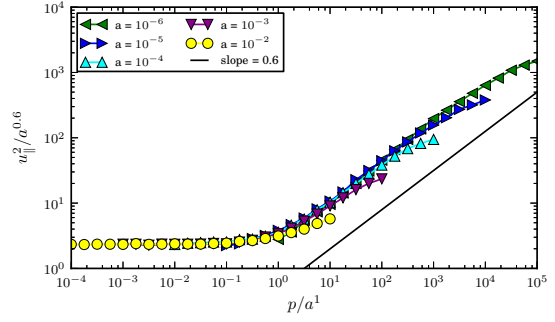
$$u_{\perp}^2 \sim a^{\lambda'' - \beta'' \nu''} p^{\beta''} \quad (4.31)$$

$$(4.32)$$

where the prime symbols are meant to distinguish the exponents from the previously used symbols for G .

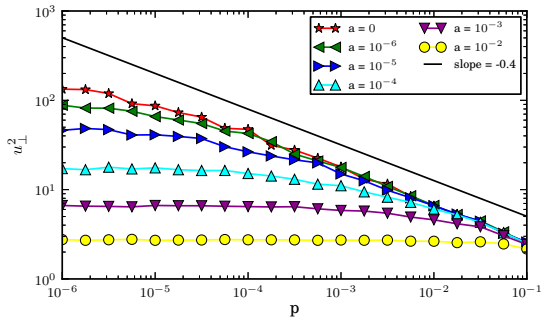


(a) Relative displacements parallel to the line of contact, $u_{\parallel}$

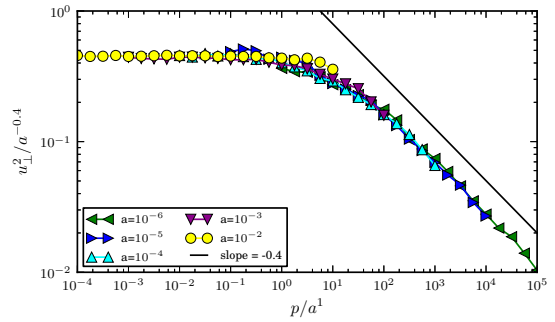


(b) Scaled values of the parallel relative displacement, $u_{\parallel}$ with attraction strength a

Figure 4.2: The parallel relative displacement, $u_{\parallel}$ is obtained from assuming a linear response to a shear strained packing. This is done for a range of attraction strengths a and pressure p . The scaled value of this displacement with a is also shown.



(a) Relative displacement perpendicular to the line of contact, $u_{\perp}$



(b) Scaled values of the perpendicular relative displacement, $u_{\perp}$ with attraction strength a

Figure 4.3: The perpendicular relative displacement, $u_{\perp}$ is obtained from assuming a linear response to a shear strained packing. This is done for a range of attraction strengths a and pressure p . The scaled value of this displacement with a is also shown.

From the theoretical scaling of the relative displacements in the case of repulsive particles, we find that :

$$\beta' = \frac{1}{2} \quad (4.33)$$

$$\lambda' = \frac{1}{2}\nu' \quad (4.34)$$

$$\beta'' = -\frac{1}{2} \quad (4.35)$$

$$\lambda'' = -\frac{1}{2}\nu'' \quad (4.36)$$

The median value of the relative displacements of all contacts from the *strain test* for the packings is shown in figure 4.2a and figure 4.3a. They are reported in terms of the squared values since the relative displacements can have both positive and negative signs. The median value is preferred here for relative displacements over the mean as there are a small number of relative displacements for contacts that skew the mean value of the data due to a large fluctuation in their values. We observe that for the case of repulsive systems :

$$u_{\parallel}^2 \sim p^{0.6} \quad (4.37)$$

$$u_{\perp}^2 \sim p^{-0.4} \quad (4.38)$$

This differs from the theoretical scaling for repulsive systems - shown previously by Wyart et al. [19] who $\Delta\phi$ used as the measure for distance to jamming - by a small amount. At the jamming transition, when the pressure is low, $u_{\perp}$ is easier to undertake for contacts than $u_{\parallel}$ so as to avoid an increase in energy change due to a reduced distance between the centres of the particles. While this holds true in the results presented in this section, the data tells us that $u_{\perp}$ doesn't increase as much as we would expect from the theoretical scaling of $u_{\perp}$ with p . And similarly for $u_{\parallel}$, when we reduce the distance to the jamming transition (i.e. $p \rightarrow 0$), the data tells us that $u_{\parallel}$ decreases more than we would expect from the theoretical scaling.

To satisfy the conditions imposed by the theoretical scaling of the relative displacements for $a = 0$ in equation 4.37, we would require $\lambda'_t = 1/2$ and $\lambda''_t = -1/2$, using the same scaling exponent for the pressure as in the case of the shear modulus, i.e. $\nu', \nu'' = 1$. The subscript t denotes the theoretical prediction for the scaling exponents. The scaled values for attractive systems is shown in figure 4.2b and figure 4.3b. From the data, we find that $\lambda' = 0.6$ and $\lambda'' = -0.4$. We get the following scaling for relative displacements :

$$\frac{u_{\parallel}^2}{a^{0.6}} \sim \left(\frac{p}{a^1}\right)^{0.6} \quad (4.39)$$

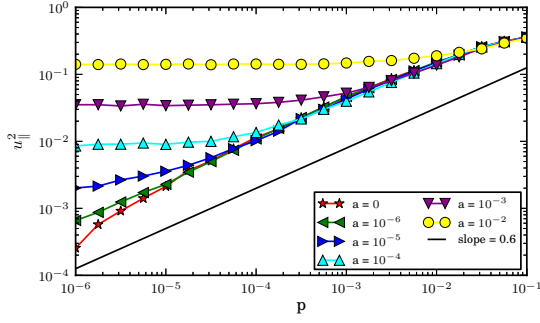
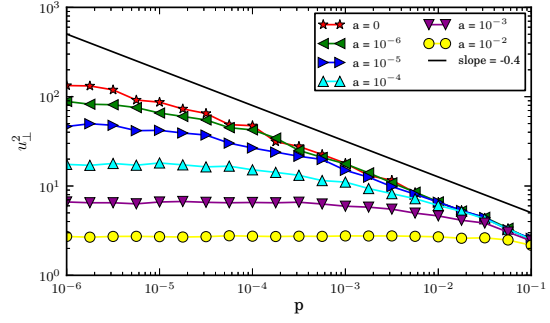
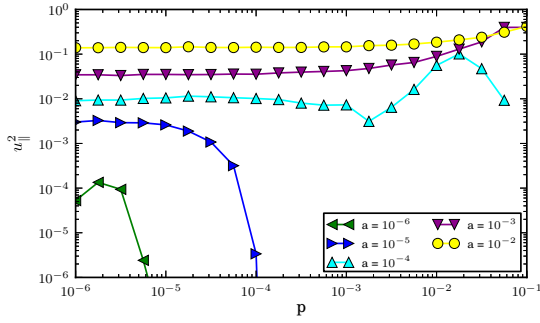
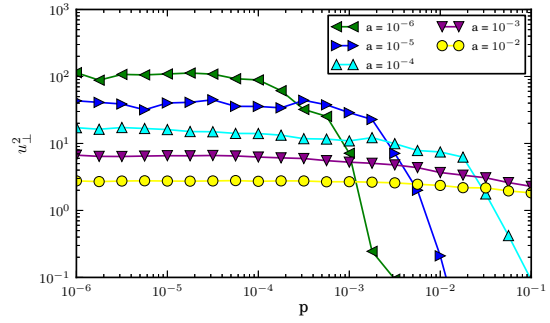
$$\frac{u_{\perp}^2}{a^{-0.4}} \sim \left(\frac{p}{a^1}\right)^{-0.4} \quad (4.40)$$

and their behaviour can be written as :

$$u_{\parallel}^2 \sim \begin{cases} a^{0.6} & \text{if, } \frac{p}{a} \ll 1 \\ p^{0.6} & \text{if, } \frac{p}{a} \gg 1 \end{cases} \quad (4.41)$$

$$u_{\perp}^2 \sim \begin{cases} a^{-0.4} & \text{if, } \frac{p}{a} \ll 1 \\ p^{-0.4} & \text{if, } \frac{p}{a} \gg 1 \end{cases} \quad (4.42)$$

In the attractive systems, two different types of contacts can be found – tensile contacts and compressive contacts. Tensile contacts are defined as those contacts where $r_{ij}/R_{ij} > 1$.

(a) Parallel, relative displacement $u_{\parallel}$, for compressive contacts(b) Perpendicular, relative displacement $u_{\perp}$, for compressive contactsFigure 4.4: Relative displacements for a range of pressure p , and varying attraction strength a for compressive contacts.(a) Parallel, relative displacement $u_{\parallel}$, for tensile contacts(b) Perpendicular, relative displacement $u_{\perp}$, for tensile contactsFigure 4.5: Relative displacements for a range of pressure p , and varying attraction strength a for tensile contacts.

This means that the force that results at such a contact is $F_{ij} < 0$; which means the particles have an attractive force between them. Similarly, compressive contacts are defined as those contacts where $r_{ij}/R_{ij} \leq 1$, which results in a contact that has a repulsive force between the particles, i.e. $F_{ij} > 0$. An obvious question that comes to mind is if these contacts behave differently in the linear response regime.

The relative displacements for tensile contacts ($u_{\parallel,t}^2, u_{\perp,t}^2$) are shown in figure 4.5a and figure 4.5b, while the relative displacements for compressive contacts ($u_{\parallel,c}^2, u_{\perp,c}^2$) are shown in figure 4.4a and figure 4.4b. At higher pressures, we find that $u_{\parallel,t}$ and $u_{\perp,t}$ decrease significantly as compared to the relative displacements of the repulsive case. The median values for the two types of contacts, tend to become similar at lower pressures. This indicates that the presence of attraction at higher pressures decreases relative displacements of tensile contacts when compared to the case of compressive contacts. The combination of attraction and low pressures doesn't seem to significantly deviate the behaviour of the relative displacements of tensile contacts when compared to compressive contacts.

Region	k_{ij}	F_0^{ij}
A	> 0	> 0
B	> 0	< 0
C	< 0	< 0

Table 4.1: Tabulated signs of the stiffness and force of a contact given for the different regions that exist in the attractive potential as shown in figure 4.6.

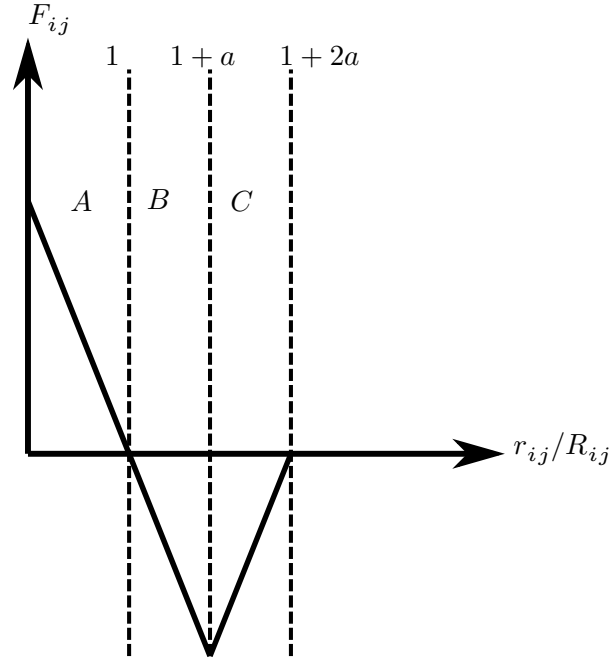


Figure 4.6: Regions with differing signs of force and stiffness of a contact that are a consequence of attractive tail of the potential that we have used.

4.4. Energy Contributions and their Scaling

We refer back to the equations we had written for a change in elastic energy when two particles in contact are displaced from their initial positions in 3 :

$$\Delta E_{ij} = \left(\frac{1}{2} k_{ij} u_{ij\parallel}^2 - \frac{1}{2r_{ij}} F_0^{ij} u_{ij\perp}^2 \right) \quad (4.43)$$

In addition to the relative displacements, we see that the stiffness k_{ij} of a contact, and the initial force F_0^{ij} between particles in contact can also change signs. This behaviour is shown in figure 4.6 and tabulated in 4.1 . As we have seen in section 4.2, the shear modulus of a packing is directly dependent on how the change in energy occurs due to the displacements of the particles. The change in energy is relatively straightforward in repulsive systems, but we see from above, it occurs differently in attractive systems. In order to quantify this difference, we look at how the mean contribution of each of the terms in table 4.1, changes with the strength of the attraction and with pressure. For a contact ij in region A , we can write the contributions to the change in elastic energy in the following way :

$$[\Delta E_{ij}]_{k,A} = \frac{1}{2} [k_{ij}]_A u_{ij\parallel}^2 \quad (4.44)$$

$$[\Delta E_{ij}]_{F,A} = -\frac{1}{2r_{ij}} [F_{ij}^0]_A u_{ij\perp}^2 \quad (4.45)$$

where the subscripts k, A and F, A represent the elastic energy contribution due to the terms associated with the stiffness constant, k_{ij} and force, F_{ij}^0 for a contact present in region A . This is done for the other terms in 4.1 as well. The following ratios are defined to look at how the different terms contribute to the mean change in energy of a contact :

$$E_I = \left\langle \frac{|\Delta E_{ij}|_{k,A/B}}{|\Delta E_{ij}|_{k,A/B} + |\Delta E_{ij}|_{F,A/B}} \right\rangle \quad (4.46)$$

$$E_{II} = \left\langle \frac{|\Delta E_{ij}|_{F,A}}{|\Delta E_{ij}|_{k,A} + |\Delta E_{ij}|_{F,A}} \right\rangle \quad (4.47)$$

$$E_{III} = \left\langle \frac{|\Delta E_{ij}|_{F,B}}{|\Delta E_{ij}|_{k,B} + |\Delta E_{ij}|_{F,B}} \right\rangle \quad (4.48)$$

$$E_{IV} = \left\langle \frac{|\Delta E_{ij}|_{k,C}}{|\Delta E_{ij}|_{k,C} + |\Delta E_{ij}|_{F,C}} \right\rangle \quad (4.49)$$

$$E_V = \left\langle \frac{|\Delta E_{ij}|_{F,C}}{|\Delta E_{ij}|_{k,C} + |\Delta E_{ij}|_{F,C}} \right\rangle \quad (4.50)$$

They are shown in figure 4.7. For the repulsive case, the relevant ratios are E_I and E_{II} , as shown in figures figure 4.7a and figure 4.7b. We see that the energy contribution of the E_I is always greater than E_2 . This is in agreement with the result that we find for relative displacements in section 4.3. The role of attraction is to increase the contribution of E_I , and decrease that of E_{II}

at low pressures. In addition, E_{III} increases at lower pressures, while E_{IV} and E_{V} have no mean contribution to the change in energy of a contact. This means that $u_{\parallel}$ is favourable compared to $u_{\perp}$. At low pressures, the particles are far enough that coming closer together by $u_{\parallel}$ will decrease the overall energy of the packing, due to the presence of attraction. We see that the pressure at which the transition to undertaking $u_{\parallel}$ is higher when the attraction strength is higher. This is also consistent with our finding for the relative displacement of contacts in section 4.3. We also see from figure 4.7d that no contacts exist in region C . This is due to the packing generation protocol that is used to make the packings. It is further explained in section 4.5.

The scaled ratios are shown in figure 4.8. We find that the energy ratios scale approximately as :

$$E_{\text{I}} \sim \begin{cases} \approx 0.8 & \text{if, } \frac{p}{a} \ll 1 \\ \approx 0.65 & \text{if, } \frac{p}{a} \gg 1 \end{cases} \quad (4.51)$$

$$E_{\text{II}} \sim \begin{cases} \approx 0.10 & \text{if, } \frac{p}{a} \ll 1 \\ \approx 0.35 & \text{if, } \frac{p}{a} \gg 1 \end{cases} \quad (4.52)$$

$$E_{\text{III}} \sim \begin{cases} \approx 0.10 & \text{if, } \frac{p}{a} \ll 1 \\ \approx 0 & \text{if, } \frac{p}{a} \gg 1 \end{cases} \quad (4.53)$$

4.5. Discussion

The first interesting observation from the previous data, is that the scaling of the pressure is always with attraction strength, p/a . This can be explained by assuming that the pressure is a function of attraction i.e. $p(a)$. When we Taylor expand this term, and divide the expansion by a , we get the scaling of pressure in attractive packings as :

$$\frac{p(a)}{a} \sim \frac{p}{a} \quad (4.54)$$

For an ordered solid, affine predictions work well for explaining the behaviour of the material under a vanishingly small shear strain. In disordered materials, this affine assumption breaks down [10, 18], and its predictions fail to explain the behaviour of the material. For a linearly shear strained packings of disordered particles, we need a framework for understanding the non-affine behaviour of the system [19]. Disordered particles, when sheared, move in two ways - they can move parallel to the line of contact, or perpendicular to it. They express their non-affine particle responses through these displacements which then leads to the divergence of the shear modulus when we close the distance to (un)jamming[19]. We can also see this in 4.9.

The study of linear response also gives us a way to connect the local properties of particles to the bulk properties of the packings, for vanishingly small shear strains. We studied in section 4.3 how these displacements change with the introduction of attraction in jammed packings, and in section 4.2, the effect of attraction on the shear modulus in the linear response regime. We saw that the shear modulus, and the relative displacements plateau when attraction is present between particles, close to the transitional jamming point (i.e. $p \rightarrow 0$ as we have used pressure as the distance to jamming). Through the energy contributions we have also seen a way to verify the scaling for the relative displacements which we obtain through the strain test.

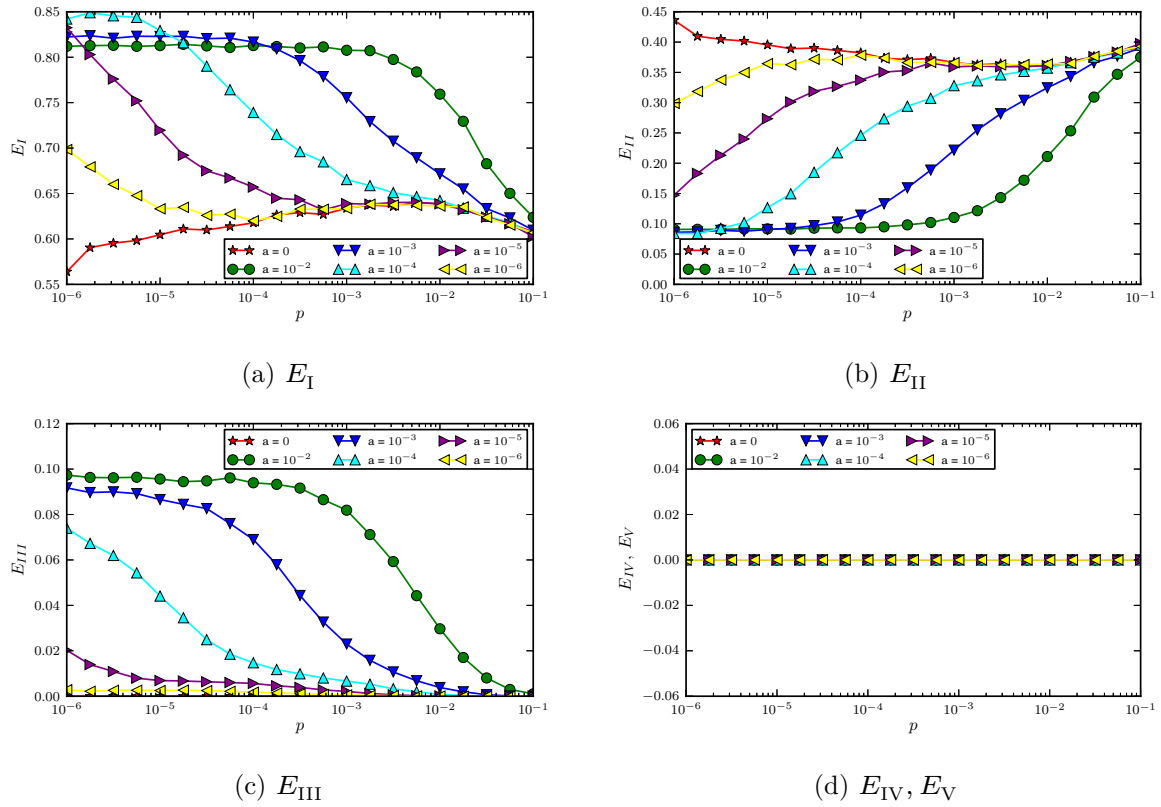


Figure 4.7: The mean contribution of various terms that show up for the change in energy of a contact, for different regions in figure 4.6 is shown here. They are defined in equation 4.51 . The scaling is done with the value of the attraction strength a .

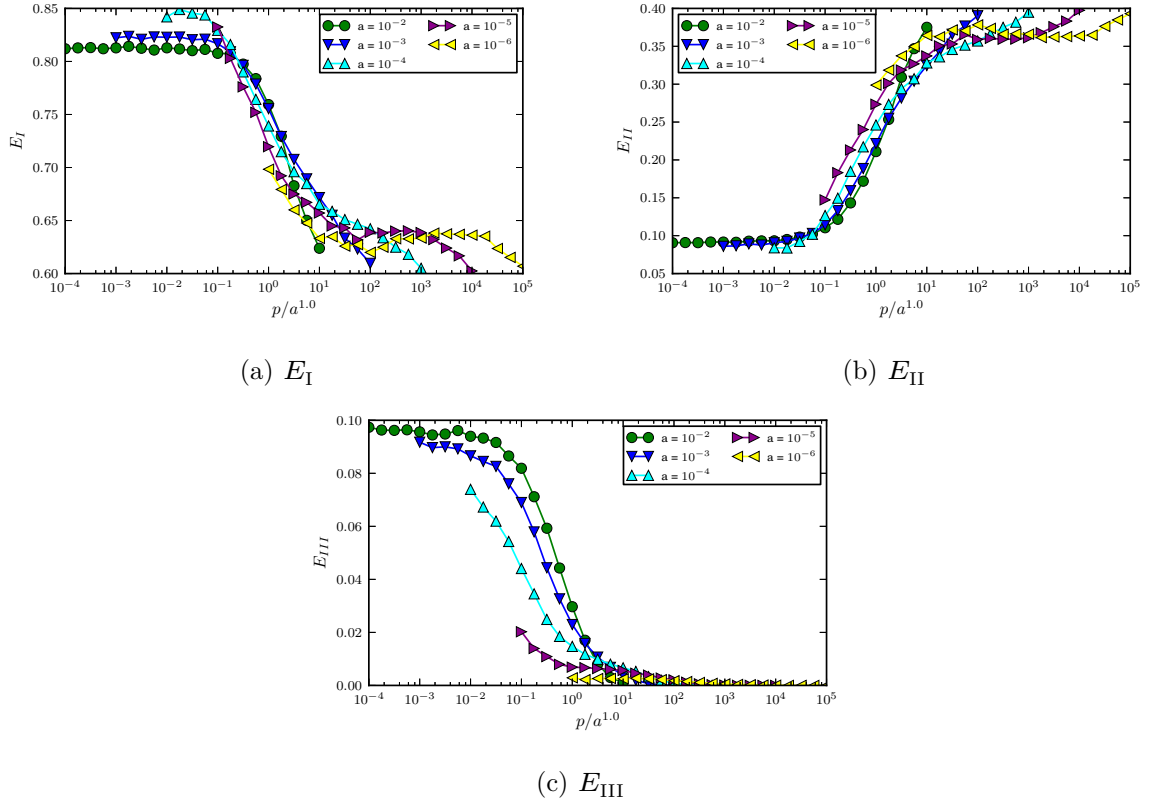


Figure 4.8: The scaled values of the average contribution of different terms that occur in the change in energy of a contact is shown here. The scaling is done with the value of the attraction strength a .

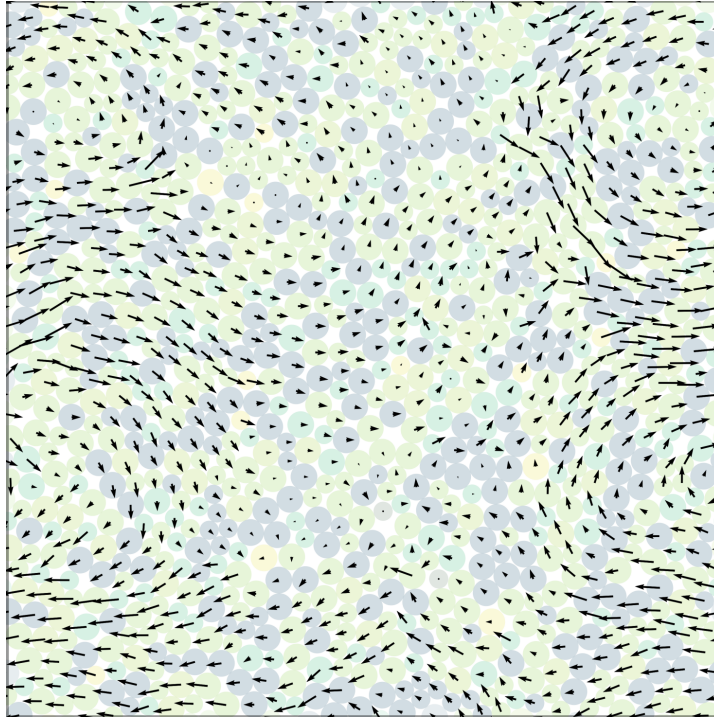


Figure 4.9: The displacements seen in a jammed, disordered packing (1024 particles, $\phi = 0.84$) when they are calculated for a soft (low energy) mode. As a way to ensure that the Hessian was working correctly in the code used for this work, we applied two verification tests. The first was to ensure that all the eigenvalues were above zero, since the Hessian matrix is a positive definite matrix (see the derivation in chapter 3). The second test, arguably the more interesting one, involved plotting the eigenvectors for one of the low energy modes (i.e. the eigenvalue that corresponds to the energy of the packing was close to $\approx 10^{-16}$ which due to finite precision of computers is essentially zero). The packing had a finite shear modulus, but the displacements of the particles turns out to be highly non-affine. This tells us exactly why when we come close to the jamming transition the value of the shear modulus diverges, at least in soft particles, the canonical model for jamming.

We have studied the linear response of packings with and without the presence of weakly attractive forces between soft spheres, using pressure as our distance to the traditional jamming point ϕ_c . In the next chapter, we shall look at the cause of the plateau values that we see in the shear modulus and the relative displacements. While we expect that weak attractive potential would have an effect on shear modulus and displacements, the plateauing of the value was quite surprising.

5

Weak Attraction and Linear Response

We had seen in section 4.4 that there was no energy contribution from contacts in region C of figure 4.6. It turns out that the weakly attractive potential used in this work, makes the protocol with which packings are formed highly protocol dependent. We measure how the pressure and the contact number of the packings for the previous chapter vary due to attraction in the following sections.

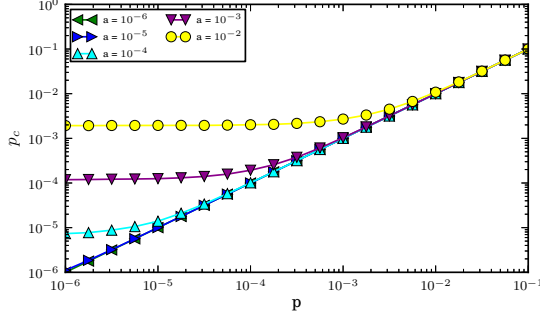
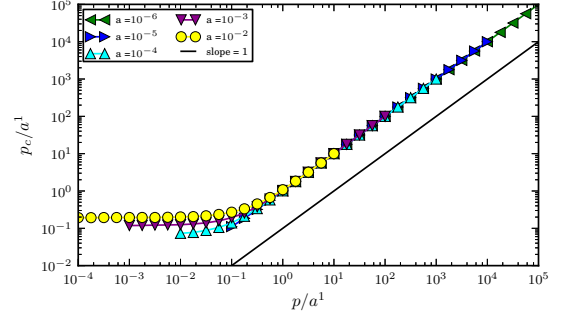
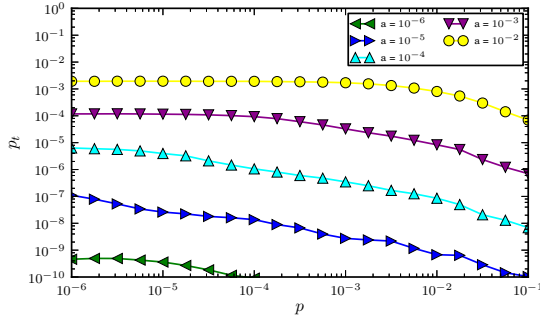
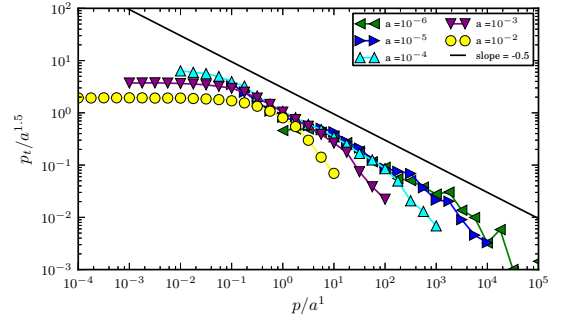
Pressure

There are two types of pressures present in attractive packings as a result of two different types of contacts – compressive and tensile pressures. They are denoted using p_c and p_t respectively. Using equation 1.7 and 1.8, we can write them as :

$$p_c = \frac{\sum_{\alpha} \sum_c F_{\alpha}^c r_{\alpha}^c}{2L^2} \quad (5.1)$$

$$p_t = \left| \frac{\sum_{\alpha} \sum_t F_{\alpha}^t r_{\alpha}^t}{2L^2} \right| \quad (5.2)$$

where α is the direction, c denotes all the compressive contacts ij between particles i and j , and t denotes all the tensile contacts. Compressive contacts would be those that exist in region A, and tensile contacts would be those that exist in region B, as shown in figure 4.6. Compressive pressure will always have a positive contribution to the total pressure, while tensile pressure will have a negative contribution to the total pressure. In order to compare the two, we use the absolute value for tensile pressure. The total pressure of a packing would be $p = p_c - p_t$. They are shown in figure 5.1a and figure 5.2a. In order to see their relative magnitudes, we plot the ratios $p_c/(p_c + p_t)$ and $p_t/(p_c + p_t)$. This is shown in 5.3. We find that the magnitude of p_c and p_t plateaus at a certain pressure. The plateauing begins to occur at a lower total pressure p for lower values of attraction. While the total pressure p decreases, we find that p_c and p_t are higher in magnitude than p .

(a) Compressive pressure p_c (b) The scaled value of the compressive pressure p_c Figure 5.1: We have shown here how the compressive pressure p_c varies with total pressure p , and the scaled value with total pressure p and attraction strength a .(a) Tensile pressure p_t (b) The scaled value of the tensile pressure p_t Figure 5.2: We have shown here how the tensile pressure p_t varies with total pressure p , and the scaled value with pressure p and attraction strength a .

The scaling for the two pressures are as follows :

$$p_c \sim \begin{cases} a & \text{if, } \frac{p}{a} \ll 1 \\ p & \text{if, } \frac{p}{a} \gg 1 \end{cases} \quad (5.3)$$

$$p_t \sim \begin{cases} a^{1.5} & \text{if, } \frac{p}{a} \ll 1 \\ pa^2 & \text{if, } \frac{p}{a} \gg 1 \end{cases} \quad (5.4)$$

Contact Number

In a similar manner to the pressure, we can also define Z_c , Z_t , the compressive and tensile contact numbers respectively, where the only change is that the type of contact that is counted for Z_c is compressive contacts, and for Z_t is tensile contacts. The total contact number Z , can be found by $Z = Z_c + Z_t$. They are shown in figure 5.4a. The individual contribution of Z_c and Z_t to Z is shown in 5.4b. We observe a similar trend for the contact number, as for pressure. In marginal jamming of a packing, we are also interested in $\Delta Z = Z - Z_{iso} = Z - 4$. We recall that excess contact number shows a jump, like pressure at the traditional jamming point and so for soft, repulsive particles it scales as $\Delta Z \sim p^{1/2}$. This is shown in figure 5.4c,

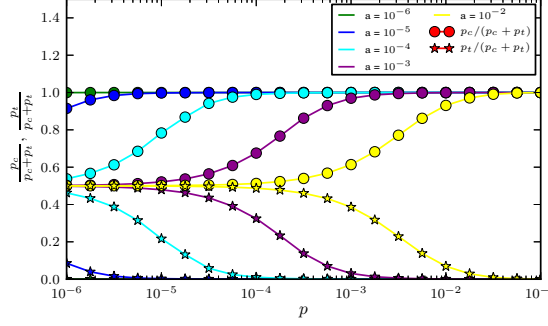


Figure 5.3: We compare, here, the relative magnitude of the compressive pressure p_c and tensile pressure p_t with to the term $p_c + p_t$

and its scaled value with attraction is shown in figure 5.4d. We find that ΔZ scales in the following way :

$$\frac{\Delta Z}{a^{0.6}} \sim \left(\frac{p}{a}\right)^{0.6} \quad (5.5)$$

$$\Delta Z \sim \begin{cases} a^{0.6} & \text{if, } \frac{p}{a} \ll 1 \\ p^{0.6} & \text{if, } \frac{p}{a} \gg 1 \end{cases} \quad (5.6)$$

$$(5.7)$$

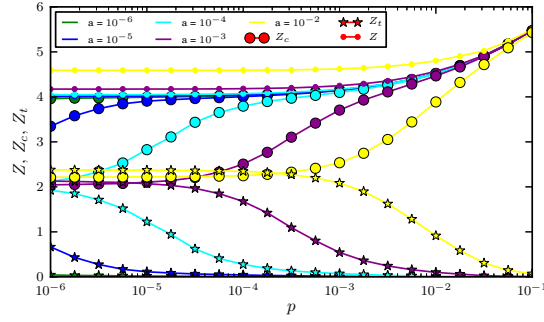
For the case of $p/a \gg 1$, we note that the theoretical scaling of Wyart et al. [19] would have been $\Delta Z \sim p^{0.5}$, while we get from our numerical simulations, $\Delta Z \sim p^{0.6}$. We checked from our numerical experiments that this also holds true for the case of zero attraction, and that $\Delta Z \sim p^{0.6}$ for the case of $a = 0$.

5.1. Discussion

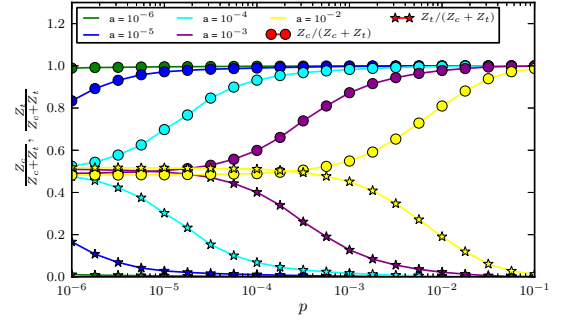
In section 5, we see that the compressive and attractive pressures *equalize* with each other at a certain pressure. By this, we mean that they come extremely close in value to each other, but never really cancelling each other out completely. The point where they equalize, is dependent on the attraction strength as we see from their scaled values. The higher the attraction strength, the higher will be the pressure, at which p_c and p_t equalize. To achieve lower pressures, after equalizing, the two pressures change by small amounts in order to give even smaller values for total pressure, p . This means that we could approach extremely low pressures with high values of attraction, without really close to what we could think of as being *marginally* jammed as in the case of soft, repulsive particles.

The plateauing observed in the displacements and shear modulus at low pressures for attractive packings, is a consequence of this *equalization* of p_c and p_t at higher pressures, due to the weakly attractive potential that exists between particles. The presence of attraction prevents packings from truly achieving low pressures, by this we mean that the packings are formed by a sort of deception by p_c and p_t , as we explained before.

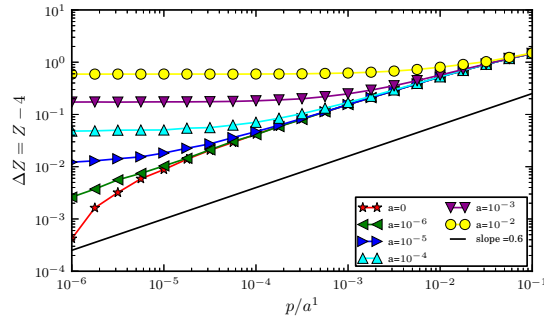
This is a consequence of the packing generation protocol we used. The packings start off from a packing fraction of $\phi = 0.9$ and the packings then are either increased or decreased in size to achieve a certain pressure. This means that the particles are very close to each other at the very beginning of the generation protocol as we have begun at a high packing fraction where the overlaps between particles is high.



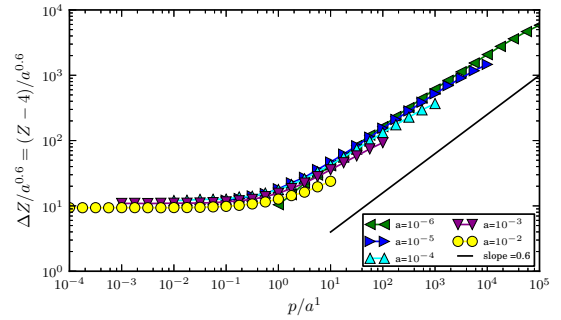
(a) The contact number Z , the compressive contact number Z_c , and the tensile contact number Z_t



(b) The contribution of Z_c, Z_t to Z



(c) Excess contact number ΔZ



(d) The scaled value of excess contact number ΔZ

Figure 5.4: We look at how the contact number ΔZ changes with attraction. The contact number has a contribution from contacts that are compressive and tensile, and their contributions, the excess contact number ΔZ and its scaled value with attraction are also shown.

This affects the regions of the potential that we can achieve between particles. This is clearly shown in section 4.4. The energy contributions of contacts in region C shown in 4.6 is zero. The contacts that we find tend to remain in regions A and B . In the potential for attractive particles, shown in 2.3, we can think of the contacts in the packings as being stuck in the potential well that exists in the corresponding regions of A and B . It is infeasible for the particles to move to region C when we increase the packing size during the packing generation protocol, since it would lead to an increase in energy of the contact between particles.

The above explanation quite accurately predicts the consequences of the packing generation protocol used when weakly attractive particles are present. This also tells us why exactly we observe plateaus in the shear modulus and relative displacements, instead of another kind of curve, that may have been possible if we used a different packing generation protocol.

In the next chapter, we shall look at the nonlinear response to *finitely* shear straining packings.

6

Results : Non Linear Response

In the previous chapters, we studied the linear response of the systems like emulsions and foams in the limit of a vanishing strain on packings. The linear response regime gives us a powerful tool to study the bulk properties of disordered systems near the jamming transition and linking it to the non affine behaviour of particles that constitute the studied systems.

An important question to ask here is what happens to packings that are close to the jamming transition, when a *finite* shear strain is applied at the boundaries of the packings. Recall that in chapter 1 we had talked about ketchup and the jammed and unjammed behaviour that it shows when we shake the bottle to make it flow more easily. In an industrial setting, knowing the mechanics that governs the behaviour of systems like emulsions and foams could lead to the setting up of efficient production processes. This is the motivation to study the effects of finite shear strain on models that describe emulsions and foams, in the context of the jamming transition. Linear elastic behaviour for materials is given by Hooke's law:

$$\sigma = G\gamma$$

and linear elasticity describes the effects of shear strain on packings when the strain is close to the vanishing strain limit quite well. We note that this was also the constitutive relationship that we had used in chapter 3. What happens at finite strain?

Boschan et al. [1] have described the behaviour of packings when we apply finite shear strain at varying strain rates to the packings. We restrict ourselves to the observations that they describe and their analysis for strain rates that are vanishingly small ($\dot{\gamma} \rightarrow 0$). This is called the quasi-static shear strain rate or the quasi-static straining action as we call it in this report.

When does linear elasticity break down and give way to the nonlinear response of packings close to the jamming transition ? It has been shown that the linear elasticity response of packings breaks down only after a small finite strain window for a quasi-static shear straining process [1]. Boschan et al. [1] observed that the measured stress with respect to the applied finite shear strain during the quasi-static shearing action followed linear elasticity (Hooke's law) up to a certain strain and then the measured stress decreased with higher values of strain. They called this strain softening, because of the decrease (or softening) in the measured stress after a certain strain.

We note that applying a finite shear strain to packings is inherently nonlinear because of the making and breaking of contacts that would take place during the shearing action, thus introducing non-linearity in the bulk response of the jammed packings. However, an interesting observation by Boschan et al. [1] was that the non linear response regime only begins after a

certain accumulation of contact changes. They were able to generalise the results obtained by van Deen et al. [17] for the strain at which the first contact change is observed in shear strained packings.

6.1. Strain Softening

In this section we will describe the quasi-static shear straining of packings for weakly attractive particles and emulsions. We generate 200 packings for a range of attraction from $a = 0, \dots, 10^{-2}$ and pressures $p = 10^{-5}, \dots, 10^{-2}$, with both pressure and attraction strength taken with a step size of a decade and by using the FCG algorithm to minimize energy, starting from an initial packing fraction of $\phi = 0.9$. Further, we impose a quasi-static, finite shear strain on these packings. In order to do so, we take start from zero strain, and subsequently vary the step size taken for shear strain $\Delta\gamma$ to reach a finite shear strain of 10^{-2} . The initial strain step that is taken is $\Delta\gamma = 10^{-6}$, and then the step size $\Delta\gamma$ is reduced further at subsequent shear strains. This is because at lower total strain we don't expect a lot of rearrangements to happen in the packings, while at higher strains, the possibility of this happening is larger.

The quasi-static shearing action, described previously takes place by applying the shear strain reached after every strain step, at the x face of the y boundaries of these packings. The particles at the boundaries are then made to comply with the shear strain by applying the Lees-Edwards boundary condition as described in chapter 4.. At each strain step, after imposing this boundary condition, we minimize the energy of the sheared packing using the FCG algorithm. This enables the sheared packing to again reach its local energy minimum after being shear strained, thus mimicking a quasi-static shear straining process.

At each shear strain value γ , we measure the shear stress of the packing using the stress tensor (as derived in appendix A). In order to observe strain softening, we plot the value of σ/γ against shear strain. This is called the relaxation shear modulus, and for clarity it'll be specified henceforth as σ/γ so as to remind the reader that it is distinct from the shear modulus that is measured in the linear response of vanishing shear strain that was studied in the previous chapter.

We show our results in figure 6.1 and figure 6.2. In figure 6.1, we show how the value of σ/γ evolves with γ for the case of packings that have a varying pressure, p , for a number of values for a , the attraction strength. In figure 6.2, we show how σ/γ evolves with γ for packings that have varying values of attraction strength, a , for a number of number of values of initial pressure p , of the packing. We have taken the median value for σ/γ at each strain step. This is necessitated by the presence of a few packings that have had such a drastic rearrangement at the beginning of the straining action that they can no longer be considered equivalent to the packing that we began with. We observe that at a certain pressure the initial value of σ/γ is higher for a higher value of a , the attraction strength. The *crossover strain* at which we observe softening in the value of σ/γ is referred to as γ_s . We observe that it increases with the the attraction strength. Therefore, the presence of attraction delays the onset of softening, and this delay is longer for a higher value of attraction strength. In addition, for the same attraction strength, packings with a higher initial pressure will undergo strain softening at a higher value of softening strain γ_s .

The role of the attraction strength and initial pressure on this crossover from linear elastic to nonlinear regime is captured by γ_s . We assume that strain softening begins at a strain where the initial value of σ/γ decreases by 70 percent. Then, we plot γ_s for various attraction strengths against initial pressure in figure 6.3a. For the case of soft, repulsive particles, it has been found that the crossover strain found by the quasi-static straining action occurs with the following scaling [1, 17] :

$$\gamma_s \sim p \quad (6.1)$$

We find that this relationship holds in figure 6.3a for $a = 0$. We scale p and γ_s , with values of attraction, and using the critical analysis technique of the previous chapter, we can write the following :

$$\frac{\gamma_s}{a^\lambda} \sim \left(\frac{p}{a^\nu}\right)^\beta \quad (6.2)$$

$$\gamma_s \sim a^{\lambda-\beta\nu} p^\beta \quad (6.3)$$

For this to hold true in the case of $a = 0$, we would require $\beta_t = 1$, and $\lambda_t = \nu_t$, where the subscript t denotes the case for theoretical prediction. From our numerical data, we find that $\beta = 0.75$, while $\lambda = 0.75$, and $\nu = 1$ i.e. $\beta = \beta_t$, while $\lambda < \nu$, which differs from our theoretical prediction of $\lambda_t = \nu_t$. The behaviour of γ_s with p and a can be written as:

$$\gamma_s \sim \begin{cases} a^{0.75} & \text{if, } \frac{p}{a} \ll 1 \\ pa^{-0.25} & \text{if, } \frac{p}{a} \gg 1 \end{cases} \quad (6.4)$$

The scaling of γ_s distinctly changes in the presence of weakly attractive forces between particles, in comparison with what we would expect from the theoretical scaling in the case of $p/a \gg 1$. In the next section, we will look at how individual particles respond to a quasi-static shear strain.

6.2. Correlation Softening

Now that we have seen how the bulk measurement σ/γ behaves during the quasi-static shearing action; we move on to how individual particles behave during this process. In order to quantify their behaviour, we need to look at their trajectories while they are quasi-statically sheared. We define a particle velocity, so as to look at how the trajectories evolve with shear strain, $\mathbf{v}_i$ for a particle i as:

$$\mathbf{v}_i = \frac{d\mathbf{x}_i}{d\dot{\gamma}} \quad (6.5)$$

where $\mathbf{x}_i$ is the particle position and $\dot{\gamma}$ is the shear shearing rate. Since we look at packings that have been quasi-statically sheared, it becomes impossible to calculate $\mathbf{v}_i$ as $\dot{\gamma} \rightarrow 0$ for this case. In order to quantify the change trajectory of the individual particles, we use the linear response of these particles to a vanishingly small shear strain. The displacements $\mathbf{q} = \{u_{1x}, u_{1y}, \dots, u_{Nx}, u_{Ny}\}$, which we get in chapter 3 as a result of linear response of particles to an infinitesimally small shear strain, gives us a way to quantify the trajectories of these particles.

The implicit assumption here is that packings are linearly elastic for vanishingly small shear strains even when we calculate trajectories from the linear response at finite strains where the linearly elasticity for a packing doesn't hold for any longer due to softening. It is based on the fact that the potential energy of the particles in quasi-statically sheared packings is still a smooth function of their displacements at any given finite strain. We link this back to chapter 3, where we had replaced contacts with springs. We have assumed that all the contacts in our quasi-statically sheared packings are in the range of separation that lie in region A and B as shown in figure 4.6. We had seen previously that no contacts lie in region C , so we can safely say here that because of the packing generation protocol used, and the fact that our packings are above the traditional jamming point when we quasi-statically shear packings, we would never see contacts that are precisely at the point where the slope of the force law changes signs (which would make an assumption of linear elasticity invalid), thus making our implicit assumption for linear elasticity at vanishingly small strains valid.

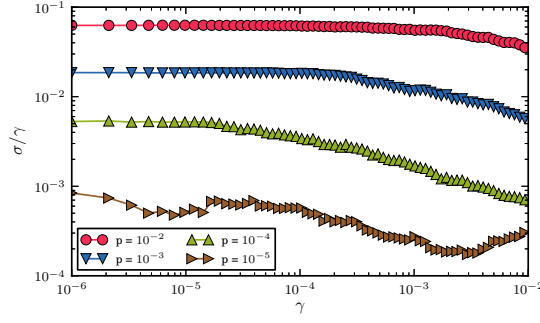
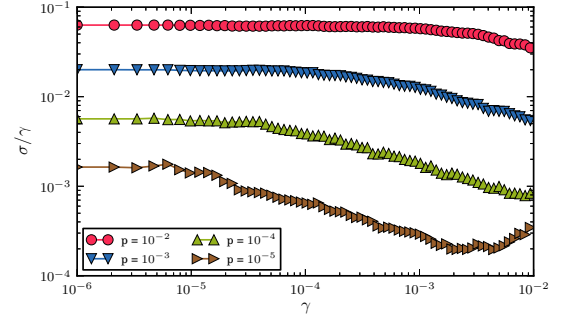
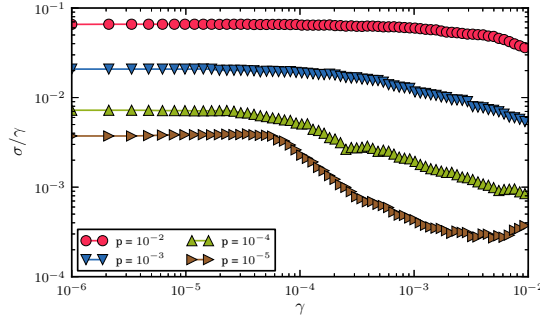
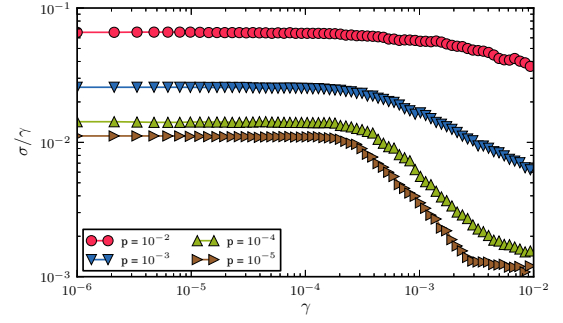
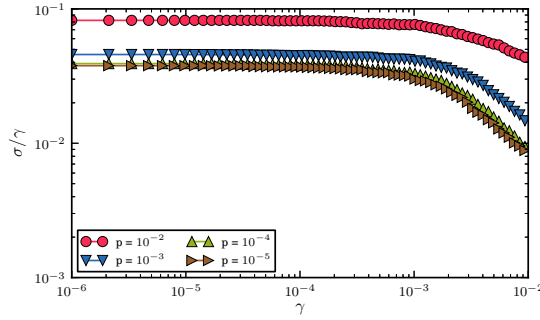
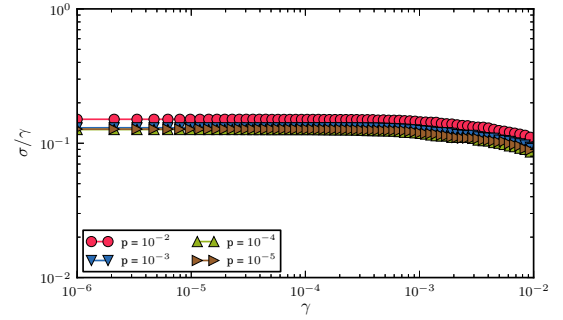
(a) $a = 0$ (b) $a = 10^{-6}$ (c) $a = 10^{-5}$ (d) $a = 10^{-4}$ (e) $a = 10^{-3}$ (f) $a = 10^{-2}$

Figure 6.1: The relaxation shear modulus $G = \sigma/\gamma$ is shown here for a number of attraction strengths. For each a , we plot the median value of σ/γ , which is found by quasi-statically shear straining packings, for a number of different pressure values p .

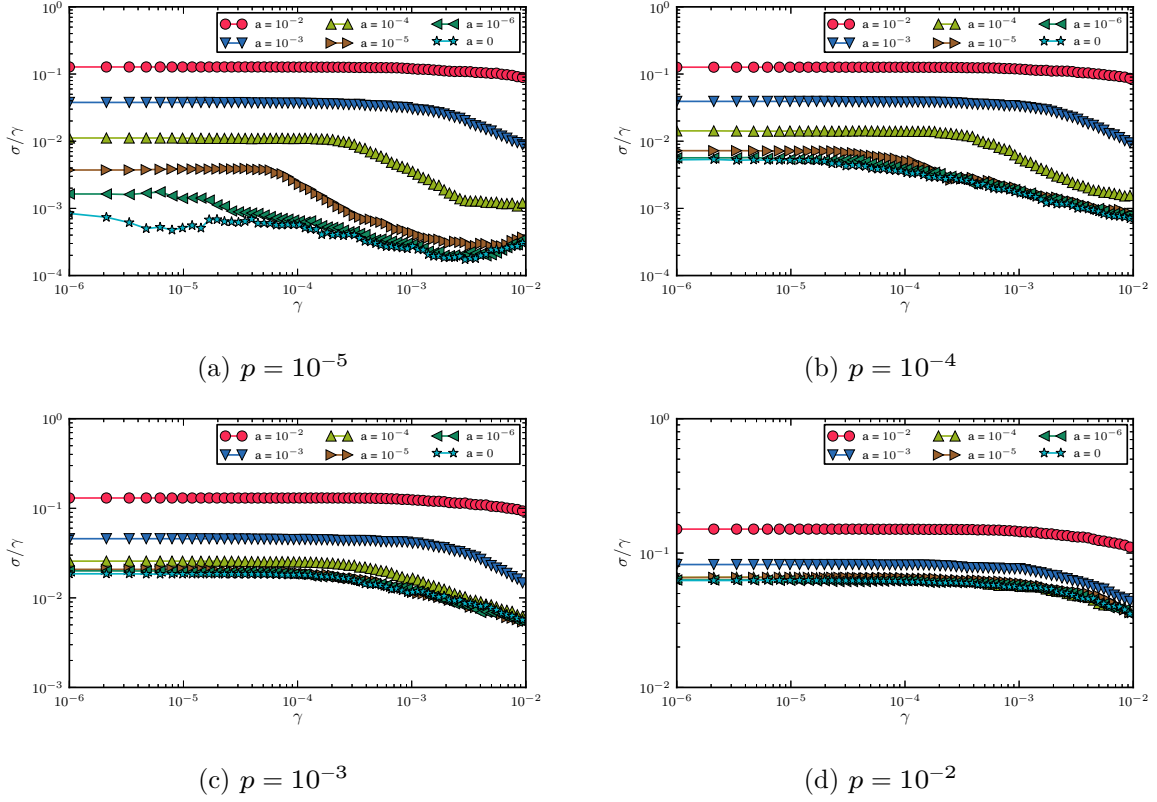


Figure 6.2: The relaxation shear modulus $G = \sigma/\gamma$ is shown here for a number of different initial pressures p of the packings. For each p , we plot the median value of σ/γ , which is found by quasi-statically shear straining packings, for a number of different attraction strengths a .

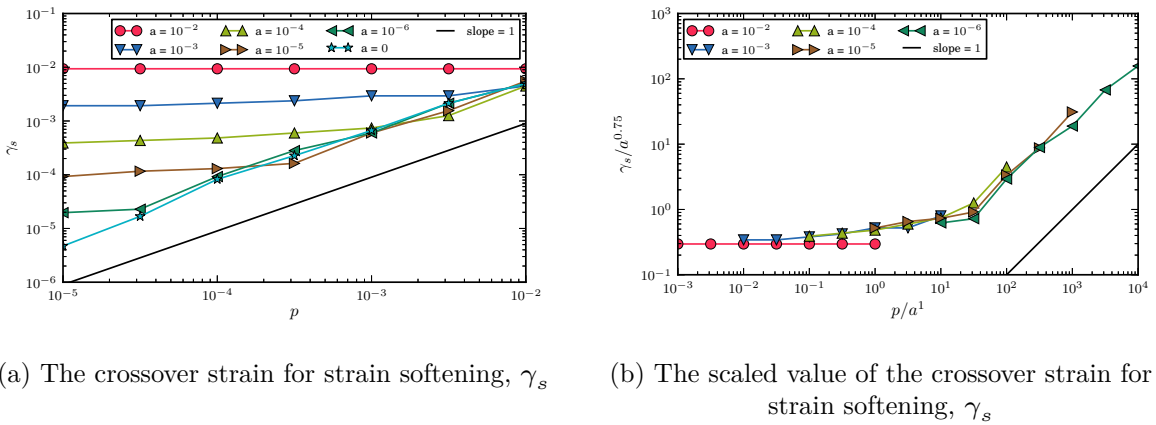


Figure 6.3: The crossover strain for strain softening γ_s as a function of initial pressure p and attraction strength a is shown here along with its scaled value.

From the previous chapter, we recall that the shear modulus of packings diverges near the jamming transition, and that the non-affinity of the relative particle displacements near the jamming transition was responsible for the divergence of the shear modulus. We see that in packings with attraction, the degree to which the shear modulus diverges is clearly dependent on both pressure and attraction strength. So, the question we would like to answer here is how dependent is the softening that we observe in the relaxation shear modulus σ/γ , which is obtained by quasi-statically shear straining packings by finite shear strain steps, on the degree of non-affinity of particle trajectories.

We begin by defining a correlation function C for these trajectories as:

$$C(\gamma) = \frac{\langle u_{iy}(\gamma) \cdot u_{iy}(0) \rangle_{i=1\dots N}}{\langle u_{iy}(0) \cdot u_{iy}(0) \rangle_{i=1\dots N}} \quad (6.6)$$

where the trajectories $u_{iy}(\gamma)$ are the displacements that we find using the strain test from the previous chapter. The subscript in $u_{iy}(\gamma)$ denotes that we only use the displacements for particles that are in the y direction. The correlation function C is measuring how trajectories vary at each strain step with their initial trajectory. We restrict ourselves to the y components of these trajectories to ensure that we only measure the non-affine components of the particle trajectories. As our shear strain is in the x direction, there would be affine and non-affine components in the displacements that take place in the x direction. Even when the trajectories begin decorrelating, and therefore showing non-affinity in their trajectories due to finite shear strain, we wouldn't be able to see it at the precise point where non-affinity in trajectories begins due to the presence of affine components in the correlation function for x components of the particle trajectories.

The median value of the correlation function, C , is shown for varying strengths of attraction for a range of pressures in figure 6.4. We find that the correlation function also shows a decrease from its initial value starting at a certain strain, similar to the way we see strain softening in section 6.1. This decrease is essentially the start of the decorrelation of the particle trajectories as discussed before; the non-affinity in particle trajectories begins to significantly increase at this point. We will call this phenomenon as *correlation softening*. The word *softening* here just means the beginning of non-affinity of particle trajectories, the choice of the word being inspired from the softening in the relaxation modulus $G = \sigma/\gamma$ observed in quasi-statically sheared packings. The crossover strain at which we observe correlation softening is called as $\gamma_s^\dagger$. In a similar manner as for the previous figure, we show in figure 6.5 how correlation softening for varying pressures and a range of attraction strengths begins. We can conclude from both these plots that the presence of attraction delays the onset of correlation softening, with the onset dependent upon the attraction strength. The initial pressure of the packing also affects the onset of correlation softening; the higher the initial pressure, the more delay there is in observing the correlation softening.

Using the same criterion as for γ_s , we assume that the crossover strain, $\gamma_s^\dagger$ for correlation softening happens when C is 70 percent of its initial value. These values are plotted against the initial pressure, for a range of attraction in 6.6a. For the case of zero attraction, we find that :

$$\gamma_s^\dagger \sim p^{0.75} \quad (6.7)$$

We do a similar critical scaling analysis, as for γ_s , and from figure 6.6b, we find that :

$$\gamma_s^\dagger \sim \begin{cases} a^{0.75} & \text{if, } \frac{p}{a} \ll 1 \\ p^{0.75} & \text{if, } \frac{p}{a} \gg 1 \end{cases} \quad (6.8)$$

6.3. Discussion

Let's assume that the presence of attraction makes it increasingly difficult for contacts to be broken when we shear packings using the protocol in section 6.1. We can make another reasonable assumption that the crossover strain from correlation softening $\gamma_s^\dagger$, which is derived from trajectories of particles in the linear response, would give us an increasingly convergent prediction for γ_s . As the attraction strength increases, the linear elastic window discussed in 6.1 should mimic the correlation C when it comes to the crossover strain due. This can be explained by the fact that the system becomes more affine when there is an increase in difficulty of breaking bonds. In order to validate this assumption, we compare the values of γ_s and $\gamma_s^\dagger$ in the two distinct regions of $p/a \ll 1$ and $p/a \gg 1$. When attraction is present, we find that :

$$\gamma_s \sim \gamma_s^\dagger \sim a^{0.75} \quad \text{if } \frac{p}{a} \ll 1 \quad (6.9)$$

and :

$$\gamma_s \sim pa^{-0.25} \quad \text{if } \frac{p}{a} \ll 1 \quad (6.10)$$

$$\gamma_s^\dagger \sim p^{0.75} \quad \text{if } \frac{p}{a} \gg 1 \quad (6.11)$$

We plot value of $\gamma_s/\gamma_s^\dagger$ in figure 6.7, and scale the pressure with attraction. We find that:

$$\frac{\gamma_s}{\gamma_s^\dagger} \sim \begin{cases} 1 & \text{if } \frac{p}{a} \ll 1 \\ \left(\frac{p}{a}\right)^{0.25} & \text{if } \frac{p}{a} \gg 1 \end{cases} \quad (6.12)$$

While γ_s and $\gamma_s^\dagger$ scale similarly when $p/a \ll 1$, our initial assumption, made at the beginning of our discussion, breaks down for the case when $p/a \gg 1$. This is because the softening observed in the correlation function has no dependence on attraction for $p/a \gg 1$ while the crossover strain for strain softening does indeed have a dependence on a (see equations 6.8 and 6.4). This means that while non-affinity increases when we apply the quasi-static straining action onto packings for $p/a \gg 1$, the crossover strain $\gamma_s^\dagger$ where the breakdown of velocity correlation begins does not depend on attraction strength. At the same time, the cross over strain for strain softening, or where the relaxation shear modulus $G = \sigma/\gamma$ begins to leave the linear elasticity regime, depends on the attraction strength.

In the case of no attraction between particles, we see that the crossover strain, for softening in the relaxation modulus, scales as $\gamma_s \sim p$ while the crossover strain, for the beginning of the breakdown in affinity of particle velocities, scales as $\gamma_s^\dagger \sim p^{3/4}$. This means that non-affinity of the particles already begins at a strain of $\gamma_s^\dagger$, before strain softening is observed, and yet the linear elasticity holds for the quasi-statically sheared packings till σ/γ softens at γ_s . This is an interesting observation when compared to what we see in repulsive particles at the jamming transition, as discussed in chapter 1, where the non-affinity of the relative particle displacements is the reason for the divergence of the shear modulus close to the jamming point J . We can conclude that non-affinity of particle velocities for quasi-statically shear strained packings does not fully explain the emergence of the softening in σ/γ or in other words the window of linear elasticity that exists at the very beginning of the quasi-static shearing action, quite surprisingly continues to exist even after non-affinity in particle velocities, derived from linear response, has set in for jammed packings.

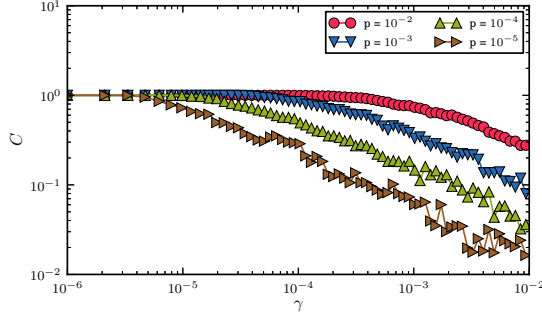
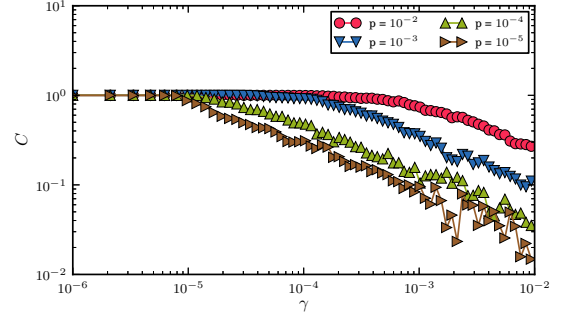
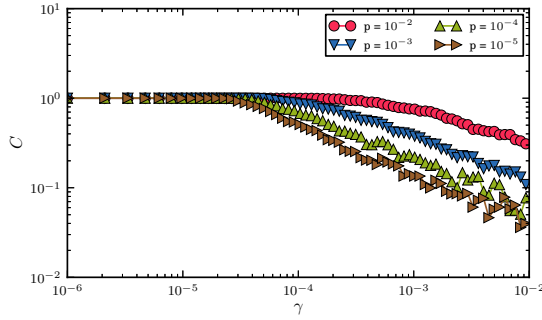
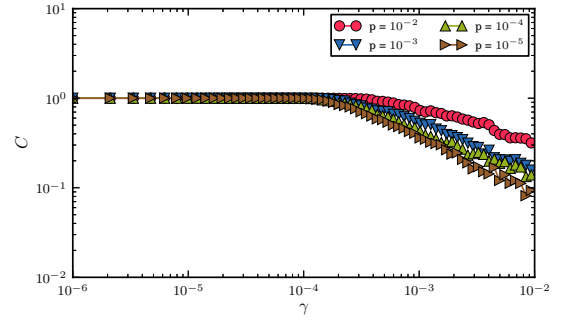
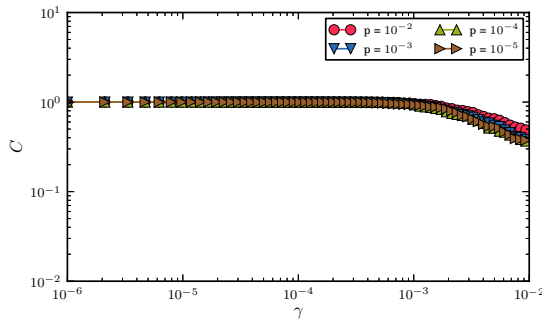
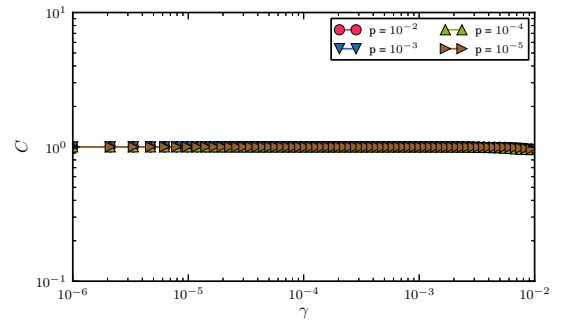
(a) $a = 0$ (b) $a = 10^{-6}$ (c) $a = 10^{-5}$ (d) $a = 10^{-4}$ (e) $a = 10^{-3}$ (f) $a = 10^{-2}$

Figure 6.4: Correlation C and its evolution with strain γ for various strengths of attraction a when quasi-statically shear straining packings is shown here.

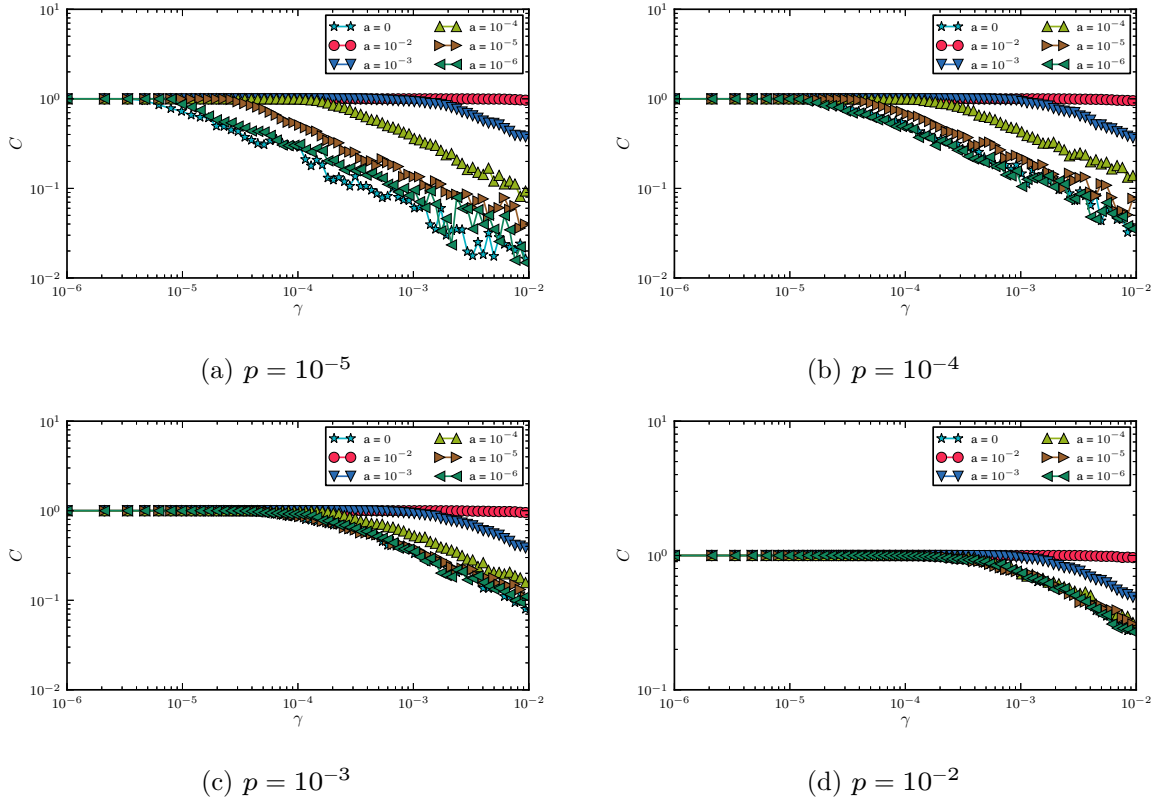


Figure 6.5: Correlation C and its evolution with strain γ for various values of the initial pressure of packing p when quasi-statically shear straining packings is shown here.

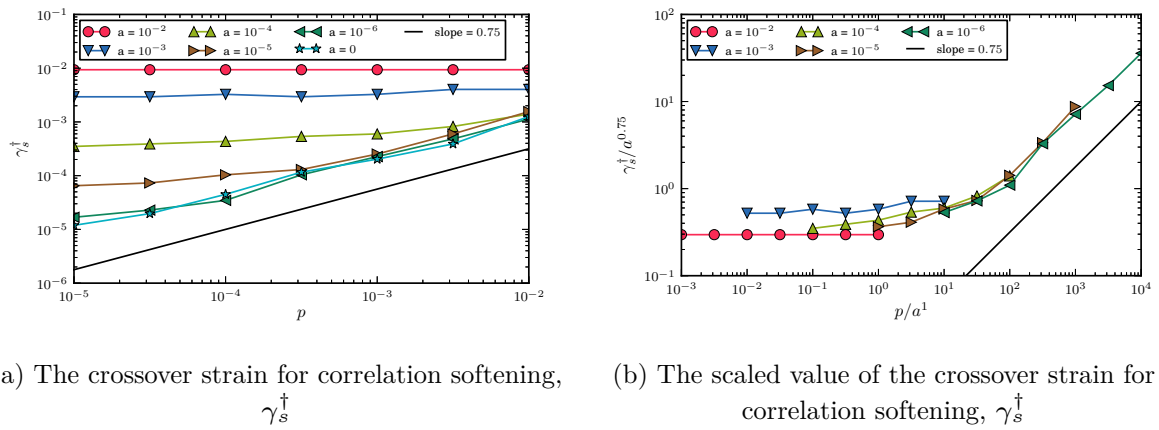


Figure 6.6: The crossover strain for correlation softening $\gamma_s^\dagger$ as a function of initial pressure p and attraction strength a is shown here along with its scaled value.

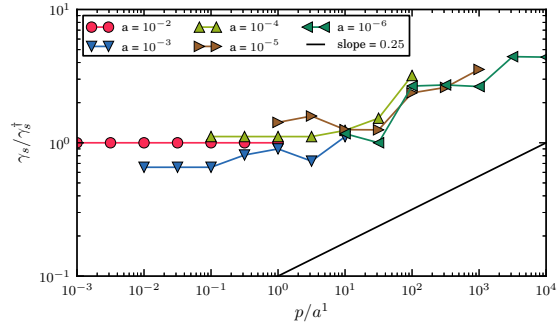


Figure 6.7: The relationship between γ_s and $\gamma_s^\dagger$ is shown here as $\gamma_s / \gamma_s^\dagger$. This value collapses onto a curve for p/a .

7

Conclusions and Future Work

We have studied how weakly attractive particles behave in the linear response regime, above the traditional critical jamming point of ϕ_c . As we have seen, the linear response regime helps in relating the non-affine character of particle motions near the marginally jammed state to bulk quantities. In the presence of attraction and for what could be considered to be traditionally jammed solids, we find the scaling relations for the shear modulus and relative displacements, using pressure as our measure the distance to the jamming point J . The plateauing of these quantities close to $p = 0$, which is our measure for the distance to the traditional jamming point ϕ_c , is explained by the packing generation protocol used to make our packings, and the potential that is used to model weakly attractive soft particles. This is verified when we look at the relative magnitudes of the compressive and tensile pressures, and the different contributions of the compressive and tensile contact number to the total contact number Z . We also looked at how the mean energy contributions of terms that make up the change in energy of a contact in the linear response, scales with pressure and attraction strength. This also gave us an understanding of the regions of the potential where the contacts in our packings could be found.

The non linear response of quasi-statically shear strained packings is also studied. We obtained two strain scales - γ_s for strain softening and $\gamma_s^\dagger$ for correlation softening - in the presence of attraction for traditionally jammed packings. The linear elastic window observed before the onset of strain softening has been a subject of vivid discussion recently, with the main discussion point being on how and why strain softening occurs [1, 15, 17]. Boschan et al. [1] argue that non linearity sets in after a set number of accumulation for contact changes that occur before γ_s , and they identified the cross over strain scale for strain softening to occur which explains the first part of this question. Therefore, we focused on the second part of this question for both weakly attractive and soft particles.

The original thesis for this work, was that the presence of attraction would prevent contact changes that occur during the quasi-static shearing action, and we could then see that the onset of non linear response at γ_s , or the closing of the linear elastic window, would correspond to a softening of a quantity derived from the linear response regime. The assumption here being that with attraction the contact network of the particles would be closer to the model of springs, in which we cannot make or break contacts, used in the linear response regime. With this in mind, we defined a correlation C , which we obtained from velocities of particles derived from the linear response at each strain step for γ . The softening of the correlation gave us a strain scale $\gamma_s^\dagger$. The softening here corresponds to the breakdown in correlation of the particle velocities obtained from the linear response at each strain step. We observe that these scales only match for the case of $p/a \ll 1$, and don't match for the case of repulsive particles

and $p/a \gg 1$. We also compared how these scales differed from one another. The fact that non-affinity in particle motions at the traditional jamming transition explains the divergence of shear modulus, but fails to completely explain the strain softening or the softening in the relaxation shear modulus seen in quasi-statically shear strained packings, points to an exciting possibility of a different mechanism at play during the finite shear straining process for both soft particles and for weakly attractive particles that model foams and emulsions. It would be useful to explicitly compare results for the relaxation shear modulus from the nonlinear response and the shear modulus obtained from linear response, in order to have a complete description of differences in bulk and particle quantities between the nonlinear response to finite shear strain and the linear response to vanishingly small shear strain.

We could also see how linear response changes when we change the protocol of our packing generation. There is a protocol dependence for the generation of packings with weakly attractive particles. It may be possible to make packings that are above ϕ_c and who also have different behaviour in the linear response due to the way the packing was made. It would be extremely interesting to study packings made below the traditional jamming point of ϕ_c . This essentially means the traditional jamming point needs to be explored further. The anomaly between the scaling obtained for relative displacements, pressure and excess contact number in our results for the linear response regime for packings made with using p as measure for the distance to jamming and what would be usually expected from well described power laws and scaling in the jamming transition literature needs to be explored further.

A

Stress Tensor Derivation

Assume that a force density of $\mathcal{F}_\alpha$ acts at the boundary of a volume $\mathcal{V}$. Let's assume a stress tensor for this volume as σ . We can then write the following using both surface and volume integrals :

$$\int_{\mathcal{V}} \mathcal{F}_\alpha d\mathcal{V} = \int_{\mathcal{V}} \frac{\partial \sigma_{\alpha\beta}}{\partial x_\beta} d\mathcal{V} \quad (\text{A.1})$$

$$\oint_{\partial\mathcal{V}} \mathcal{F}_\alpha d\mathcal{A} = \oint_{\partial\mathcal{V}} \sigma_{\alpha\beta} n_\beta d\mathcal{A} \quad (\text{A.2})$$

We can also write this as :

$$\frac{\partial(x_\gamma \sigma_{\alpha\beta})}{\partial x_\alpha} = \frac{\partial x_\gamma}{\partial x_\alpha} \sigma_{\alpha\beta} + x_\gamma \frac{\partial \sigma_{\alpha\beta}}{\partial x_\alpha} \quad (\text{A.3})$$

$$= \delta_{\alpha\gamma} \sigma_{\alpha\beta} + x_\gamma \frac{\partial \sigma_{\alpha\beta}}{\partial x_\alpha} \quad (\text{A.4})$$

For stress balance we require that :

$$x_\gamma \frac{\partial \sigma_{\alpha\beta}}{\partial x_\alpha} = 0 \quad (\text{A.5})$$

Therefore :

$$\frac{\partial(x_\gamma \sigma_{\alpha\beta})}{\partial x_\alpha} = \delta_{\alpha\gamma} \sigma_{\alpha\beta} \quad (\text{A.6})$$

$$= \sigma_{\alpha\gamma} \quad (\text{A.7})$$

This can done for the entire volume $\mathcal{V}$, note that we have used β instead of γ here for convenience :

$$\frac{1}{\mathcal{V}} \int_{\mathcal{V}} \sigma_{\alpha\beta} d\mathcal{V} = \frac{1}{\mathcal{V}} \int_{\mathcal{V}} \sigma_{\alpha\gamma} d\mathcal{V} \quad (\text{A.8})$$

$$= x_\gamma \sigma_{\alpha\beta} n_\beta d\mathcal{A} \quad (\text{A.9})$$

We define the average stress tensor $\bar{\sigma}_{\alpha\beta}$ as :

$$\bar{\sigma}_{\alpha\beta} = \frac{1}{\mathcal{V}} \int_{\mathcal{V}} \sigma_{\alpha\beta} d\mathcal{V} \quad (\text{A.10})$$

The force density can also be written in the following manner :

$$\sigma_{\alpha\beta} n_{\beta} = \mathcal{F}_{\alpha} \quad (\text{A.11})$$

Using the above, we can write :

$$\bar{\sigma}_{\alpha\beta} = \frac{1}{\mathcal{V}} \int_{\partial\mathcal{V}} x_{\gamma} \mathcal{F}_{\alpha} d\mathcal{V} \quad (\text{A.12})$$

Let's now assume that there is a volume V that is filled with N particles. By using Voronoi tessellation, we can find the volume occupied by a single particle. Let's assume that a single particle occupies a volume of $\mathcal{V}$ and it is in contact with other particles. We will call these contacts as c . The average stress tensor for this particle can be written as :

$$\bar{\sigma}_{\alpha\beta} = \frac{N}{V} \oint_{dV/N} \sum_c F_{\alpha}^c x_{\beta}^c dA \quad (\text{A.13})$$

Since these are discrete forces and their sum is finite, we can write :

$$\bar{\sigma}_{\alpha\beta} = \frac{N}{V} \sum_c F_{\alpha}^c x_{\beta}^c \quad (\text{A.14})$$

We can now find the average stress tensor for all the particles as :

$$\bar{\sigma}_{\alpha\beta} = \frac{N}{V} \sum_{\langle i,j \rangle} F_{\alpha}^{ij} x_{\beta}^{ij} \quad (\text{A.15})$$

where i and j are the particles that are in contact.

B

Packing generation protocol

Packings can be generated using the non-linear conjugate gradient method [15]. Initially, all the particles in the packing are put at random points inside of the box. The goal now is to go to a minimum possible energy for the packing. Let's assume that all the particle positions are given by $\mathbf{q}$. The energy is given by U . The forces between particles in contact are given by F . There's two ways of achieving our goal. The first is the energy gradient method (ECG), and the second is the force gradient method (FCG).

B.1. Energy Gradient Method

We need to get to a minimum in the energy landscape of our initial, randomly generated packing. The non linear conjugate gradient (NCG) algorithm is used to achieve this since at every iteration of the algorithm, the positions of the particles, and therefore the energy changes. The NCG algorithm searches for the steepest direction. It does so by using the gradient of the energy. The gradient of the energy gives us the forces $-\nabla U = \mathbf{F}$. This is used as the first guess of the search direction in NCG. We also know that the potential between two particles is proportional to their overlap. It gives us an estimate of how the energy curve with respect to the search direction would look like - essentially a parabola. If we have three points of a parabola, we will have enough information to construct this parabola. With the NCG, we can take three steps, with three search directions, and end up with an estimation of the parabola. The minimum point of this parabola would then give us the minimum energy.

A disadvantage of this step is that we can't be certain that we have achieved the minimum due to numerical issues. In short, because we start off with a certain energy initially we have zeroth order terms that do not allow us to reach an equilibrium of the packing to its minimum energy. The zero-th order terms cause catastrophic cancellation and therefore limits us from achieving the energy minimum. This leads us to another way of making packings and its called the Force Conjugate Gradient(FCG) method.

B.2. Force Conjugate Gradient Method

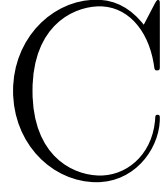
Instead of minimizing energy, we can also minimize the forces between particles in a packing. Since at a minimum energy, the gradient is zero, and therefore forces are zero, we have no zero-th order terms. This means that there is no catastrophic cancellation like the Energy gradient method. In addition, we only have to do two steps of the NCG, since we are now essentially looking for the search direction where the gradient is zero.

All of the packings are made by achieving a certain pressure. This is done by starting at a certain packing fraction, in our case $\phi = 0.9$, and randomly populating the packing. The

box size is then increased or decreased by a certain amount, and FCG is used to then get the configuration of particles which results in minimum energy. The pressure of the packing is then calculated. Depending on whether it is greater or lesser than the desired pressure, the box size is increased or decreased. And so on and so forth, the iterations take place.

B.3. Attractive Particles

In the case of attractive particles, the potential between two particles isn't as easy as the potential that we see in the case of repulsive particles. If they come really close to one another, the attractive part of the potential takes over, and the tendency of the particles is to come even closer, so as to decrease the total energy of the system. This means that the parabola that we used in the repulsive case for ECG does not exist any more, which necessitates the use of FCG.



Absolute and Relative Expressions

C.1. Relative Potential for the repulsive case

Overlap is defined as :

$$\delta_{ij} = 1 - \frac{r_{ij}}{R_{ij}} \quad (\text{C.1})$$

C.1.1. Potential

$$V_{ij} = \begin{cases} \frac{\epsilon}{2} \left(1 - \frac{r_{ij}}{R_{ij}}\right)^2, & \text{if } \frac{r_{ij}}{R_{ij}} \leq 1 \\ 0, & \text{if } \frac{r_{ij}}{R_{ij}} > 1 \end{cases} \quad (\text{C.2})$$

$$V_{ij} = \begin{cases} \frac{\epsilon}{2} \delta_{ij}^2, & \text{if } \delta_{ij} \geq 0 \\ 0, & \text{if } \delta_{ij} < 0 \end{cases} \quad (\text{C.3})$$

C.1.2. Force

$$F_{ij} = \begin{cases} \frac{\epsilon}{R_{ij}} \left(1 - \frac{r_{ij}}{R_{ij}}\right), & \text{if } \frac{r_{ij}}{R_{ij}} \leq 1 \\ 0, & \text{if } \frac{r_{ij}}{R_{ij}} > 1 \end{cases} \quad (\text{C.4})$$

$$F_{ij} = \begin{cases} \frac{\epsilon \delta_{ij}}{R_{ij}}, & \text{if } \delta_{ij} \geq 0 \\ 0, & \text{if } \delta_{ij} < 0 \end{cases} \quad (\text{C.5})$$

C.1.3. Stiffness

$$k_{ij} = \begin{cases} \frac{\epsilon}{R_{ij}^2}, & \text{if } \frac{r_{ij}}{R_{ij}} \leq 1 \\ 0, & \text{if } \frac{r_{ij}}{R_{ij}} > 1 \end{cases} \quad (\text{C.6})$$

$$k_{ij} = \begin{cases} \frac{\epsilon}{R_{ij}^2}, & \text{if } \delta_{ij} \geq 0 \\ 0, & \text{if } \delta_{ij} < 0 \end{cases} \quad (\text{C.7})$$

C.2. Relative Potential for the attractive case

Attractive overlap is defined as :

$$\delta_{ij}^{attr} = 1 + 2a - \frac{r_{ij}}{R_{ij}} \quad (C.8)$$

C.2.1. Potential

$$V_{ij} = \begin{cases} \frac{\epsilon}{2} \left[\left(1 - \frac{r_{ij}}{R_{ij}} \right)^2 - 2a^2 \right] & , \text{if } \frac{r_{ij}}{R_{ij}} \leq 1 + a \\ \frac{\epsilon}{2} \left[1 + 2a - \frac{r_{ij}}{R_{ij}} \right]^2 & , \text{if } 1 + a < \frac{r_{ij}}{R_{ij}} \leq 1 + 2a \\ 0 & , \text{if } \frac{r_{ij}}{R_{ij}} > 1 + 2a \end{cases} \quad (C.9)$$

$$V_{ij} = \begin{cases} \frac{\epsilon}{2} (\delta_{ij}^2 - 2a^2) & , \text{if } \delta_{ij}^{attr} > a \\ -\frac{\epsilon}{2} (\delta_{ij}^{attr})^2 & , \text{if } 0 < \delta_{ij}^{attr} < a \\ 0 & , \text{if } \delta_{ij}^{attr} < 0 \end{cases} \quad (C.10)$$

C.2.2. Force

$$F_{ij} = \begin{cases} \frac{\epsilon}{R_{ij}} \left(1 - \frac{r_{ij}}{R_{ij}} \right) & , \text{if } \frac{r_{ij}}{R_{ij}} \leq 1 + a \\ -\frac{\epsilon}{R_{ij}} \left(1 + 2a - \frac{r_{ij}}{R_{ij}} \right) & , \text{if } 1 + a < \frac{r_{ij}}{R_{ij}} \leq 1 + 2a \\ 0 & , \text{if } \frac{r_{ij}}{R_{ij}} > 1 + 2a \end{cases} \quad (C.11)$$

$$F_{ij} = \begin{cases} \frac{\epsilon \delta_{ij}}{R_{ij}} & , \text{if } \delta_{ij}^{attr} > a \\ -\frac{\epsilon \delta_{ij}^{attr}}{R_{ij}} & , \text{if } 0 < \delta_{ij}^{attr} < a \\ 0 & , \text{if } \delta_{ij}^{attr} < 0 \end{cases} \quad (C.12)$$

C.2.3. Stiffness

$$k_{ij} = \begin{cases} \frac{\epsilon}{R_{ij}^2} & , \text{if } \frac{r_{ij}}{R_{ij}} \leq 1 + a \\ -\frac{\epsilon}{R_{ij}^2} & , \text{if } 1 + a < \frac{r_{ij}}{R_{ij}} \leq 1 + 2a \\ 0 & , \text{if } \frac{r_{ij}}{R_{ij}} > 1 + 2a \end{cases} \quad (C.13)$$

$$k_{ij} = \begin{cases} \frac{\epsilon}{R_{ij}^2} & , \text{if } \delta_{ij}^{attr} > a \\ -\frac{\epsilon}{R_{ij}^2} & , \text{if } 0 < \delta_{ij}^{attr} < a \\ 0 & , \text{if } \delta_{ij}^{attr} < 0 \end{cases} \quad (C.14)$$

C.3. Absolute Potential for the repulsive case

Absolute overlap is defined as :

$$\delta_{ij}^{abs} = R_{ij} - r_{ij} \quad (C.15)$$

C.3.1. Potential

$$V_{ij} = \begin{cases} \frac{\epsilon}{2} (R_{ij} - r_{ij})^2 & , \text{if } r_{ij} \leq R_{ij} \\ 0 & , \text{if } r_{ij} > R_{ij} \end{cases} \quad (C.16)$$

$$V_{ij} = \begin{cases} \frac{\epsilon}{2} (\delta_{ij}^{abs})^2 & , \text{if } \delta_{ij}^{abs} \geq 0 \\ 0 & , \text{if } \delta_{ij}^{abs} < 0 \end{cases} \quad (C.17)$$

C.3.2. Force

$$F_{ij} = \begin{cases} \epsilon (R_{ij} - r_{ij}) & , \text{if } r_{ij} \leq R_{ij} \\ 0 & , \text{if } r_{ij} > R_{ij} \end{cases} \quad (C.18)$$

$$F_{ij} = \begin{cases} \epsilon \delta_{ij}^{abs} & , \text{if } \delta_{ij}^{abs} \geq 0 \\ 0 & , \text{if } \delta_{ij}^{abs} < 0 \end{cases} \quad (C.19)$$

C.3.3. Stiffness

$$k_{ij} = \begin{cases} \epsilon & , \text{if } r_{ij} \leq R_{ij} \\ 0 & , \text{if } r_{ij} > R_{ij} \end{cases} \quad (C.20)$$

$$k_{ij} = \begin{cases} \epsilon & , \text{if } \delta_{ij}^{abs} \geq 0 \\ 0 & , \text{if } \delta_{ij}^{abs} < 0 \end{cases} \quad (C.21)$$

C.4. Absolute Potential for the attractive case

Absolute, attractive potential is defined as :

$$\delta_{ij}^{attr,abs} = (1 + 2a)(R_{ij} - r_{ij}) \quad (C.22)$$

C.4.1. Potential

$$V_{ij} = \begin{cases} \frac{\epsilon}{2} [(R_{ij} - r_{ij})^2 - 2(R_{ij}a)^2] & , \text{if } r_{ij} \leq R_{ij}(1 + a) \\ \frac{\epsilon}{2} [(1 + 2a)R_{ij} - r_{ij}]^2 & , \text{if } R_{ij}(1 + a) < r_{ij} \leq R_{ij}(1 + 2a) \\ 0 & , \text{if } r_{ij} > R_{ij}(1 + 2a) \end{cases} \quad (C.23)$$

$$V_{ij} = \begin{cases} \frac{\epsilon}{2} [(\delta_{ij}^{abs})^2 - 2(R_{ij}a)^2] & , \text{if } \delta_{ij}^{attr,abs} \geq R_{ij}a \\ -\frac{\epsilon}{2} (\delta_{ij}^{attr,abs})^2 & , \text{if } R_{ij}a > \delta_{ij}^{attr,abs} > 0 \\ 0 & , \text{if } \delta_{ij}^{attr,abs} < 0 \end{cases} \quad (C.24)$$

C.4.2. Force

$$F_{ij} = \begin{cases} \epsilon (R_{ij} - r_{ij}) & , \text{if } r_{ij} < R_{ij}(1 + a) \\ -\epsilon (R_{ij}(1 + 2a) - r_{ij}) & , \text{if } R_{ij}(1 + a) < r_{ij} < R_{ij}(1 + 2a) \\ 0 & , \text{if } r_{ij} > R_{ij}(1 + 2a) \end{cases} \quad (\text{C.25})$$

$$F_{ij} = \begin{cases} \epsilon \delta_{ij}^{abs} & , \text{if } \delta_{ij}^{attr,abs} \geq R_{ij}a \\ -\epsilon \delta_{ij}^{attr,abs} & , \text{if } R_{ij}a > \delta_{ij}^{attr,abs} > 0 \\ 0 & , \text{if } \delta_{ij}^{attr,abs} < 0 \end{cases} \quad (\text{C.26})$$

C.4.3. Stiffness

$$k_{ij} = \begin{cases} \epsilon & , \text{if } r_{ij} < R_{ij}(1 + a) \\ -\epsilon & , \text{if } R_{ij}(1 + a) < r_{ij} < R_{ij}(1 + 2a) \\ 0 & , \text{if } r_{ij} > R_{ij}(1 + 2a) \end{cases} \quad (\text{C.27})$$

$$k_{ij} = \begin{cases} \epsilon & , \text{if } \delta_{ij}^{attr,abs} \geq R_{ij}a \\ -\epsilon & , \text{if } R_{ij}a > \delta_{ij}^{attr,abs} > 0 \\ 0 & , \text{if } \delta_{ij}^{attr,abs} < 0 \end{cases} \quad (\text{C.28})$$

D

Dynamical Matrix Expressions

D.1. Expressions for the Hessian

When all of the expressions found in chapter 3 for the relative case of repulsive potential are put into the 3.10 equation and we look at the expressions for the Hessian of all the particles in contact, we find that :

$$\frac{\partial^2}{\partial q_a^\alpha \partial q_b^\beta} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 \right) = A \frac{\epsilon}{R_{ij}^2 r_{ij}^2} \alpha_{ij} \beta_{ij} \quad (D.1)$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial q_b^\beta} \left(\frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \frac{\epsilon}{R_{ij} r_{ij}^3} \left(1 - \frac{r_{ij}}{R_{ij}} \right) (B r_{ij}^2 - A \alpha_{ij} \beta_{ij}) \quad (D.2)$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial q_b^\beta} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 - \frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \frac{\epsilon}{R_{ij}} \left(A \frac{\alpha_{ij}}{r_{ij}} \frac{\beta_{ij}}{r_{ij}} \left(\frac{1}{R_{ij}} + \frac{\delta_{ij}}{r_{ij}} \right) - B \frac{\delta_{ij}}{r_{ij}} \right) \quad (D.3)$$

where :

$$a, b = \{i, j\}, \text{ where } i \text{ and } j \text{ are a pair of interacting particles} \quad (D.4)$$

$$\alpha, \beta = \{x, y\}, \text{ where } x \text{ and } y \text{ are the cartesian basis of the absolute frame} \quad (D.5)$$

$$\alpha_{ij}, \beta_{ij} = X_{ij} \text{ or } Y_{ij} \quad (D.6)$$

$$A = \begin{cases} -1 & , \text{ if } a \neq b \\ 1 & , \text{ if } a = b \end{cases} \quad (D.7)$$

$$B = \begin{cases} -1 & , \text{ if } a \neq b \text{ and } \alpha = \beta \\ 1 & , \text{ if } a = b \text{ and } \alpha = \beta \\ 0 & , \text{ if } \alpha \neq \beta \end{cases} \quad (D.8)$$

$$\delta_{ij} = \left(1 - \frac{r_{ij}}{R_{ij}} \right) , \text{ called the overlap} \quad (D.9)$$

D.2. Box Degree of Freedom : Shear Strain

Let us assume a box degree of freedom, namely the *shear strain*, and represented by γ , that can be perturbed in addition to the particles' degrees of freedom, as talked about in chapter

3. The expressions for the new terms are as follows:

$$\frac{\partial^2}{\partial q_a^\alpha \partial \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 \right) = C \frac{\epsilon L_y}{R_{ij}^2 r_{ij}^2} X_{ij} \alpha_{ij} \quad (\text{D.10})$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial \gamma} \left(\frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = D \frac{\epsilon}{R_{ij} r_{ij}^3} L_y \delta_{ij} Y_{ij} \beta_{ij} \quad (\text{D.11})$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 - \frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \frac{\epsilon L_y}{R_{ij} r_{ij}^2} \left(C \frac{X_{ij} \alpha_{ij}}{R_{ij}} - D \frac{\delta_{ij} Y_{ij} \beta_{ij}}{r_{ij}} \right) \quad (\text{D.12})$$

$$\frac{\partial^2}{\partial^2 \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 \right) = \frac{\epsilon L_y^2}{R_{ij}^2 r_{ij}^2} X_{ij}^2 \quad (\text{D.13})$$

$$\frac{\partial^2}{\partial^2 \gamma} \left(\frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \frac{\epsilon L_y^2}{R_{ij} r_{ij}^3} \delta_{ij} Y_{ij}^2 \quad (\text{D.14})$$

$$\frac{\partial^2}{\partial^2 \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 - \frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \frac{\epsilon L_y^2}{R_{ij} r_{ij}^2} \left(\frac{X_{ij}^2}{R_{ij}} - \frac{\delta_{ij} Y_{ij}^2}{r_{ij}} \right) \quad (\text{D.15})$$

where:

$$L_y = \text{Length of box in } y \text{ direction} \quad (\text{D.16})$$

$$C = \begin{cases} 1 & , \text{ if } a = j \\ -1 & , \text{ if } a \neq j \end{cases} \quad (\text{D.17})$$

$$D = \begin{cases} -1 & , \text{ if } a \neq j, \alpha = x \text{ or } a = j, \alpha \neq x \\ 1 & , \text{ if } a = j, \alpha = x \text{ or } a \neq j, \alpha \neq x \end{cases} \quad (\text{D.18})$$

$$(\text{D.19})$$

D.3. Attractive Potential between Particles

We shall now shift our focus to particles that also have a weak attraction between them, at short range. The potential is of the following form between two particles i, j :

$$V_{ij} = \begin{cases} \frac{\epsilon}{2} \left[\left(1 - \frac{r_{ij}}{R_{ij}} \right)^2 - 2a^2 \right] & , \text{ if } \frac{r_{ij}}{R_{ij}} < 1 + a \\ -\frac{\epsilon}{2} \left[1 + 2a - \frac{r_{ij}}{R_{ij}} \right]^2 & , \text{ if } 1 + a < \frac{r_{ij}}{R_{ij}} < 1 + 2a \\ 0 & , \text{ if } \frac{r_{ij}}{R_{ij}} > 1 + 2a \end{cases} \quad (\text{D.20})$$

where a is the attraction factor that defines the strength of the attraction between the particles.

D.4. Hessian Expressions for the Attractive Potential

Using the previous notations we get the following if $\frac{r_{ij}}{R_{ij}} < 1 + a$:

$$\frac{\partial^2}{\partial q_a^\alpha \partial q_b^\beta} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 \right) = A \frac{\epsilon}{R_{ij}^2 r_{ij}^2} \alpha_{ij} \beta_{ij} \quad (D.21)$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial q_b^\beta} \left(\frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \frac{\epsilon}{R_{ij} r_{ij}^3} \left(1 - \frac{r_{ij}}{R_{ij}} \right) (B r_{ij}^2 - A \alpha_{ij} \beta_{ij}) \quad (D.22)$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial q_b^\beta} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 - \frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \frac{\epsilon}{R_{ij}} \left(A \frac{\alpha_{ij}}{r_{ij}} \frac{\beta_{ij}}{r_{ij}} \left(\frac{1}{R_{ij}} + \frac{\delta_{ij}}{r_{ij}} \right) - B \frac{\delta_{ij}}{r_{ij}} \right) \quad (D.23)$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 \right) = C \frac{\epsilon L_y}{R_{ij}^2 r_{ij}^2} X_{ij} \alpha_{ij} \quad (D.24)$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial \gamma} \left(\frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = D \frac{\epsilon}{R_{ij} r_{ij}^3} L_y \delta_{ij} Y_{ij} \beta_{ij} \quad (D.25)$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 - \frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \frac{\epsilon L_y}{R_{ij} r_{ij}^2} \left(C \frac{X_{ij} \alpha_{ij}}{R_{ij}} - D \frac{\delta_{ij} Y_{ij} \beta_{ij}}{r_{ij}} \right) \quad (D.26)$$

$$\frac{\partial^2}{\partial^2 \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 \right) = \frac{\epsilon L_y^2}{R_{ij}^2 r_{ij}^2} X_{ij}^2 \quad (D.27)$$

$$\frac{\partial^2}{\partial^2 \gamma} \left(\frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \frac{\epsilon L_y^2}{R_{ij} r_{ij}^3} \delta_{ij} Y_{ij}^2 \quad (D.28)$$

$$\frac{\partial^2}{\partial^2 \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 - \frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \frac{\epsilon L_y^2}{R_{ij} r_{ij}^2} \left(\frac{X_{ij}^2}{R_{ij}} - \frac{\delta_{ij} Y_{ij}^2}{r_{ij}} \right) \quad (D.29)$$

In the above case, it is observed that the expressions are identical to the ones that are obtained in the case of repulsive potential between the particles. The expressions change for the next case, where we see the attraction parameter coming into play. If $1 + a < \frac{r_{ij}}{R_{ij}} < 1 + 2a$, the the

expressions becomes the following,

$$\frac{\partial^2}{\partial q_a^\alpha \partial q_b^\beta} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 \right) = -A \frac{\epsilon}{R_{ij}^2 r_{ij}^2} \alpha_{ij} \beta_{ij} \quad (D.30)$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial q_b^\beta} \left(\frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = -\frac{\epsilon}{R_{ij} r_{ij}^3} \left(1 + 2a - \frac{r_{ij}}{R_{ij}} \right) (B r_{ij}^2 - A \alpha_{ij} \beta_{ij}) \quad (D.31)$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial q_b^\beta} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 - \frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \frac{\epsilon}{R_{ij}} \left(-A \frac{\alpha_{ij}}{r_{ij}} \frac{\beta_{ij}}{r_{ij}} \left(\frac{1}{R_{ij}} + \frac{\delta_{ij}^{attr}}{r_{ij}} \right) + B \frac{\delta_{ij}^{attr}}{r_{ij}} \right) \quad (D.32)$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 \right) = -C \frac{\epsilon L_y}{R_{ij}^2 r_{ij}^2} X_{ij} \alpha_{ij} \quad (D.33)$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial \gamma} \left(\frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = -D \frac{\epsilon}{R_{ij} r_{ij}^3} L_y \delta_{ij}^{attr} Y_{ij} \beta_{ij} \quad (D.34)$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 - \frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \frac{\epsilon L_y}{R_{ij} r_{ij}^2} \left(-C \frac{X_{ij} \alpha_{ij}}{R_{ij}} + D \frac{\delta_{ij}^{attr} Y_{ij} \beta_{ij}}{r_{ij}} \right) \quad (D.35)$$

$$\frac{\partial^2}{\partial^2 \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 \right) = -\frac{\epsilon L_y^2}{R_{ij}^2 r_{ij}^2} X_{ij}^2 \quad (D.36)$$

$$\frac{\partial^2}{\partial^2 \gamma} \left(\frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = -\frac{\epsilon L_y^2}{R_{ij} r_{ij}^3} \delta_{ij}^{attr} Y_{ij}^2 \quad (D.37)$$

$$\frac{\partial^2}{\partial^2 \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 - \frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = -\frac{\epsilon L_y^2}{R_{ij} r_{ij}^2} \left(\frac{X_{ij}^2}{R_{ij}} - \frac{\delta_{ij}^{attr} Y_{ij}^2}{r_{ij}} \right) \quad (D.38)$$

The variables above, are defined in the same way as before. The only new expression is :

$$\delta_{ij}^{attr} = 1 + 2a - \frac{r_{ij}}{R_{ij}}, \text{ which is called the attraction overlap} \quad (D.39)$$

D.5. Absolute Overlap for Attractive Particles

In a different scenario, where we use the potential that is not dimensionless, which is called absolute overlap, we get a slightly modified set of equations. The absolute overlap is expressed as follows:

$$V_{ij} = \begin{cases} \frac{\epsilon}{2} \left[(R_{ij} - r_{ij})^2 - 2(R_{ij}a)^2 \right] & , \text{ if } \frac{r_{ij}}{R_{ij}} < 1 + a \\ -\frac{\epsilon}{2} \left[R_{ij}(1 + 2a) - r_{ij} \right]^2 & , \text{ if } 1 + a < \frac{r_{ij}}{R_{ij}} < 1 + 2a \\ 0 & , \text{ if } \frac{r_{ij}}{R_{ij}} > 1 + 2a \end{cases} \quad (D.40)$$

We shall now define our overlaps in a different manner than before:

$$\delta_{ij} = R_{ij} - r_{ij}, \text{ which is called the absolute overlap} \quad (D.41)$$

$$\delta_{ij}^{attr} = R_{ij}(1 + 2a) - r_{ij}, \text{ which is called the absolute attractive overlap} \quad (D.42)$$

The expressions from before now are slightly modified. So if $\frac{r_{ij}}{R_{ij}} < 1 + a$, we get :

$$\frac{\partial^2}{\partial q_a^\alpha \partial q_b^\beta} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 \right) = A \frac{\epsilon}{r_{ij}^2} \alpha_{ij} \beta_{ij} \quad (D.43)$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial q_b^\beta} \left(\frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \frac{\epsilon}{r_{ij}^3} (R_{ij} - r_{ij}) (Br_{ij}^2 - A\alpha_{ij}\beta_{ij}) \quad (D.44)$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial q_b^\beta} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 - \frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \epsilon \left(A \frac{\alpha_{ij}}{r_{ij}} \frac{\beta_{ij}}{r_{ij}} \left(1 + \frac{\delta_{ij}}{r_{ij}} \right) - B \frac{\delta_{ij}}{r_{ij}} \right) \quad (D.45)$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 \right) = C \frac{\epsilon L_y}{r_{ij}^2} X_{ij} \alpha_{ij} \quad (D.46)$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial \gamma} \left(\frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = D \frac{\epsilon}{r_{ij}^3} L_y \delta_{ij} Y_{ij} \beta_{ij} \quad (D.47)$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 - \frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \frac{\epsilon L_y}{r_{ij}^2} \left(CX_{ij} \alpha_{ij} - D \frac{\delta_{ij} Y_{ij} \beta_{ij}}{r_{ij}} \right) \quad (D.48)$$

$$\frac{\partial^2}{\partial^2 \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 \right) = \frac{\epsilon L_y^2}{r_{ij}^2} X_{ij}^2 \quad (D.49)$$

$$\frac{\partial^2}{\partial^2 \gamma} \left(\frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \frac{\epsilon L_y^2}{r_{ij}^3} \delta_{ij} Y_{ij}^2 \quad (D.50)$$

$$\frac{\partial^2}{\partial^2 \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 - \frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \frac{\epsilon L_y^2}{r_{ij}^2} \left(X_{ij}^2 - \frac{\delta_{ij} Y_{ij}^2}{r_{ij}} \right) \quad (D.51)$$

In the above case, we also observe that the expressions are identical to the ones that are obtained in the case of repulsive potential between the particles. The expressions change for the next case, where we see the attraction parameter coming into play. If $1 + a < \frac{r_{ij}}{R_{ij}} < 1 + 2a$,

the the expressions becomes the following,

$$\frac{\partial^2}{\partial q_a^\alpha \partial q_b^\beta} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 \right) = -A \frac{\epsilon}{r_{ij}^2} \alpha_{ij} \beta_{ij} \quad (\text{D.52})$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial q_b^\beta} \left(\frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = -\frac{\epsilon}{r_{ij}^3} \delta_{ij}^{attr} (Br_{ij}^2 - A\alpha_{ij}\beta_{ij}) \quad (\text{D.53})$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial q_b^\beta} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 - \frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = \epsilon \left(-A \frac{\alpha_{ij}}{r_{ij}} \frac{\beta_{ij}}{r_{ij}} \left(1 + \frac{\delta_{ij}^{attr}}{r_{ij}} \right) + B \frac{\delta_{ij}^{attr}}{r_{ij}} \right) \quad (\text{D.54})$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 \right) = -C \frac{\epsilon L_y}{r_{ij}^2} X_{ij} \alpha_{ij} \quad (\text{D.55})$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial \gamma} \left(\frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = -D \frac{\epsilon}{r_{ij}^3} L_y \delta_{ij} Y_{ij} \beta_{ij} \quad (\text{D.56})$$

$$\frac{\partial^2}{\partial q_a^\alpha \partial \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 - \frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = -\frac{\epsilon L_y}{r_{ij}^2} \left(CX_{ij} \alpha_{ij} - D \frac{\delta_{ij} Y_{ij} \beta_{ij}}{r_{ij}} \right) \quad (\text{D.57})$$

$$\frac{\partial^2}{\partial^2 \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 \right) = -\frac{\epsilon L_y^2}{r_{ij}^2} X_{ij}^2 \quad (\text{D.58})$$

$$\frac{\partial^2}{\partial^2 \gamma} \left(\frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = -\frac{\epsilon L_y^2}{r_{ij}^3} \delta_{ij}^{attr} Y_{ij}^2 \quad (\text{D.59})$$

$$\frac{\partial^2}{\partial^2 \gamma} \left(\frac{k_{ij}}{2} u_{ij\parallel}^2 - \frac{F_0^{ij}}{2r_{ij}} u_{ij\perp}^2 \right) = -\frac{\epsilon L_y^2}{r_{ij}^2} \left(X_{ij}^2 - \frac{\delta_{ij}^{attr} Y_{ij}^2}{r_{ij}} \right) \quad (\text{D.60})$$

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