Dynamical Heterogeneity in Granular Fluids and Structural Glasses

A dissertation presented to
the faculty of
the College of Arts and Sciences of Ohio University

In partial fulfillment
of the requirements for the degree
Doctor of Philosophy

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May 2014

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This dissertation titled
Dynamical Heterogeneity in Granular Fluids and Structural Glasses

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ABSTRACT

AVILA, KARINA E., Ph.D., May 2014, Physics and Astronomy

Dynamical Heterogeneity in Granular Fluids and Structural Glasses (101 pp.)

Director of Dissertation: Horacio E. Castillo

Our current understanding of the dynamics of supercooled liquids and other similar slowly evolving (“glassy”) systems is rather limited. One aspect that is particularly poorly understood is the origin and behavior of the strong non trivial fluctuations that appear in the relaxation process toward equilibrium. Glassy systems and granular systems both present regions of particles moving cooperatively and at different rates from other regions. This phenomenon is known as spatially heterogeneous dynamics. A detailed explanation of this phenomenon may lead to a better understanding of the slow relaxation process, and perhaps it could even help to explain the presence of the glass transition.

This dissertation concentrates on studying dynamical heterogeneity by analyzing simulation data for models of granular materials and structural glasses.

For dissipative granular fluids, the growing behavior of dynamical heterogeneities is studied for different densities and different degrees of inelasticity in the particle collisions. The correlated regions are found to grow rapidly as the system approaches dynamical arrest. Their geometry is conserved even when probing at different cutoff length in the correlation function or when the energy dissipation in the system is increased.

For structural glasses, I test a theoretical framework that models dynamical heterogeneity as originated in the presence of Goldstone modes, which emerge from a broken continuous time reparametrization symmetry. This analysis is based on quantifying the size and the spatial correlations of fluctuations in the time variable and of other kinds of fluctuations. The results obtained here agree with the predictions of the hypothesis. In particular, the fluctuations associated to the time reparametrization invariance become stronger for low temperatures, long timescales, and large coarse graining lengths.
Overall, this research points to dynamical heterogeneity to be described for granular systems similarly than for other glassy systems and it provides evidence in favor of a particular theory for the origin of dynamical heterogeneity.
DEDICATION

to Stefan and Alan
ACKNOWLEDGMENTS

First and foremost I would like to thank Prof. Horacio E. Castillo for his guidance during the completion of this work; for his advice not only with professional concerns but also with personal questions. I would also like to thank Prof. Annette Zippelius for her friendly acceptance in her group and for all the help she provided to me. I thank Prof. Konrad Samwer for considering me as a member of his group. Also, I thank Prof. Katharina Vollmayr-Lee for very interesting discussions.

I would like to thank the Ohio University Condensed Matter and Surface Sciences Program (CMSS) for a studentship, the Ohio University Nanoscale and Quantum Phenomena Institute (NQPI) for support as a research assistant, Ohio University for support as a teaching assistant, the Max Planck Institute for Dynamics and Self-organization for a fellowship, and the United States Department of Energy for support as a research assistant under grant DE-FG02-06ER46300.

I also would like to thank the members of my thesis committee Profs. David Drabold, David Tees and Eric Masson for taking the time to evaluate my work.

Many thanks to Profs. Madappa Prakash and Daniel Phillips for their very valuable help and advice.

For their help in many administrative and technical matters I would like to thank the physics and astronomy department staff: Wayne Chiasson, Candy Dishong, Julie Goettge, Tracy Inman and Don Roth. From the University of Göttingen I thank the secretaries Gabriele Schubert and Katrin Glormann.

Special thanks to Andrea Fiege, Iraj Gholami and Till Kranz for very helpful discussions. I am also very grateful to Dr. Richard Vink for his kind support.

I would also like to thank David Ruiz for his friendship and support. To all the wonderful people I met during my stay in Ohio and Göttingen. In particular I want to
thank Mahmoud Asmar, Alice von der Heydt, Daniel Wilkin, Sina Sadeghi, Stephan Ulrich, Antje Krüger, Jon-Olaf Krisponeit and Sebastian Pitikaris. Many thanks.

I am very thankful to my family that has always been there for me. Specially, many thanks to my sister Melina Avila and to my brother in law Daniel Santiago which have been a great support in every aspect of my life. Also, as part of my family I thank Rudi and Christa Küchemann for all their help when I most needed it.

Last but not least, I would like to thank Stefan and Alan Küchemann for the wonderful time they give me in our home. Their constant love always gives me the strength to be better in everything I do.
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LIST OF ACRONYMS

PDF  Pair Distribution Function
LJ   Lennard Jones
WCA  Weeks Chandler Andersen
KWW  Kohlrausch William Watts
TTI  Time Translation Invariant
MSD  Mean Square Displacement
OZ   Ornstein Zernicke
NVT  Constant: number of particles N, volume V and temperature T
NPT  Constant: number of particles N, pressure P and temperature T
1 AN OVERVIEW OF GLASSES

1.1 Introduction

Amorphous systems have been the object of intense study for many years [1]. These materials are solid, but they do not have the spatially periodic characteristic of crystals and they are also out of equilibrium. Due to the numerous applications of glasses, a large effort has been devoted over the years to understand their properties [2]. However, our theoretical understanding of the dynamical behavior of glasses is still very limited, and it remains one of the most challenging problems in Condensed Matter Physics. A variety of models have been devised to explain glass dynamics [3–8], but most of them are only successful for very specific purposes and do not give sufficient insight into the general problem.

Glassy systems exhibit characteristics such as slow relaxation, aging (i.e., the breakdown of time translation invariance), and dynamical heterogeneities (i.e., strong fluctuations in the dynamics near the glass transition [2]). Other systems that present similar characteristics are dense granular materials and dense colloids. The study of these systems is useful for the understanding of glassy behavior, and, in contrast with structural glasses, they are accessible to imaging techniques which allow the tracking of trajectories of individual particles.

The main focus of this work is the study of dynamical heterogeneity in simulations of two different kinds of systems, dissipative granular fluids and structural glasses. In this introductory chapter I review the general characteristics of the systems as well as some basic observations about the phenomenology of amorphous material. Moreover, I give a summary of the physical quantities used in the study of dynamical heterogeneity and introduce a hypothesis explaining the origin of dynamical heterogeneities. At the end of this chapter the goal and the organization of this work are summarized.
1.2 Structural Glasses and the Glass Transition

Structural glasses are amorphous substances which exhibit the rigidity of solids. Typically, a structural glass is prepared through quenching, which consists in cooling from the liquid state very quickly below the melting temperature, $T_m$, as shown in Fig. 1.1 [9]. When the liquid passes the temperature $T_m$ without crystallizing it is called a supercooled liquid. While decreasing the temperature the motion of the molecules gets slower and slower, until they cannot rearrange into an equilibrium configuration within the time scale of the experiment. The temperature at which this happens is called the glass transition temperature [1], $T_g$. Below this temperature the system is solid but its structure resembles

![Diagram](image_url)  

**Figure 1.1:** Temperature dependence of a liquid’s specific volume $V_{sp}$. $T_m$ is the melting temperature. A fast cooling rate results in a glass transition at $T_{g1}$; a slower cooling rate leads to a glass transition $T_{g2}$. Reprinted with permission from Ref. [9]. Copyright (1996) American Chemical Society.
that of a liquid. We say that the system has become a structural glass. The value of $T_g$ depends on the cooling rate, the faster we cool the system the higher its value. Another indication of the slowing down of the dynamics is that the viscosity and relaxation time are strongly dependent on temperature and they increase dramatically as $T_g$ is approached.

### 1.3 Granular Materials and the Jamming Transition

Granular media are materials made of grains like sand, ball bearings or powders [10]. The particles are macroscopic and therefore they exhibit no thermal motion. They collide inelastically, so they rapidly come to rest unless energy is supplied to the system by shearing, shaking, or other means, in which case it is said that the system is “fluidized”. When the packing fraction $\phi$ of the fluidized granular system is sufficiently high, the particles may present dynamical arrest, and in that case the system is said to be jammed [11]. The packing fraction at which dynamical arrest occurs is called the jamming point, $\phi_J$. In a 2D granular system this point is found to be around the value $\phi_J \approx 0.82$. It is commonly believed that the stress relaxation behavior of granular materials, as well as of colloids, can be described in a similar fashion to the behavior of supercooled liquids [10]. In Ref. [11], the idea of a jamming phase diagram was presented. This diagram, shown in Fig. 1.2, shows that different systems “unjam” by varying different control parameters. Structural glasses unjam as the temperature rises, while colloids and granular materials unjam when the packing fraction is lowered. In the phase diagram, all three control parameters are important for these systems, however, their significance for each individual system varies. Whether or not the idea of a universal jamming phase diagram is correct, and in particular whether the jamming transition is equivalent to the glass transition, is still a matter of strong debate.
1.4 Structure and Dynamics of Amorphous Systems

1.4.1 Structure

The absence of long range order in glasses can be captured in the pair distribution function (PDF), $g(r)$. This function measures the probability of finding another atom at a separation $r$ from a given atom. The peaks of $g(r)$ correspond to the nearest neighbor shells. These peaks vanish as the radial distance from the origin is increased, due to the disordered structure. This is illustrated in the sketch presented in Fig. 1.3. Nevertheless, the few peaks in $g(r)$ in this illustration show that there is short range order to some degree. This means that the particles are not positioned randomly [12].
1.4.2 Non–exponential Relaxation and Aging Observed in Two–time Correlation Functions

In glasses, the relaxation behavior usually does not obey an exponential law. This is sometimes described by the Kohlraush-Williams-Watts (KWW) or stretched exponential function

\[ \Phi(t) = \exp\left[-(t/\tau)^\beta\right], \quad (1.1) \]

where \( \beta \) takes a value between 0 and 1, and \( \tau \) is a relaxation time [1]. The reason for this relaxation behavior has not been fully determined yet. A possible explanation for this phenomenon is introduced in Sec. 1.5.

This non–exponential relaxation (see Eq. 1.1) can be observed in two-time correlation functions, like the self part of the intermediate scattering function
\[ C(t, t_w) = \frac{1}{N} \sum_{i=1}^{N} \cos [\vec{k} \cdot (\vec{r}_i(t) - \vec{r}_i(t_w))], \tag{1.2} \]

where \( i \) is the index for the particle, \( N \) is the number of particles, \( \vec{r}_i(t_w) \) the position of particle \( i \) at the “waiting time” \( t_w \), \( \vec{r}_i(t) \) the position of particle \( i \) at the “final time” \( t \) (with \( t_w < t \)) and \( \vec{k} \) is a fixed wavevector. The decay of \( C(t, t_w) \) as a function of \( t \), for fixed \( t_w \) (Fig. 1.4) provides information about the mobility of the particles between the two given times \( t_w \) and \( t \). A value of \( C(t, t_w) \) near unity indicates that the system has not changed significantly between the two times, while a value of \( C(t, t_w) \) near zero indicates that the system has changed dramatically. The curves show a characteristic pattern called “two step relaxation”. The first step is a relaxation to a plateau with value \( q_{EA} \) (the Edwards-Anderson order parameter) when the particles are trapped in cages which are formed by their neighbors. This is followed by a much slower relaxation called “alpha relaxation”, characterized by the relaxation time \( \tau_\alpha \), which is related to diffusion after the breaking of the cages. The alpha relaxation time is defined by the condition
\[ C(t_w + \tau_\alpha(t_w), t_w) = 1/e. \]

Another possible definition of the correlation function \( C(t, t_w) \) can be
\[ C_a(t, t_w) = \frac{1}{N} \sum_{i=1}^{N} \theta(a - |\vec{r}_i(t) - \vec{r}_i(t_w)|), \tag{1.3} \]

where \( \theta = 1 \) if \( |\vec{r}_i(t) - \vec{r}_i(t_w)| \leq a \), or zero otherwise. This quantity behaves qualitatively in a similar way to the one defined in Eq. (1.2). Note that in order to distinguish this new quantity from Eq. (1.2), the subindex \( a \) has been introduced in the notation.

In equilibrium, and more generally in time translationally invariant (TTI) systems, Eq. (1.2) depends only on the difference between the two times \( t - t_w \). In the case of aging, which is another characteristic of amorphous systems, the system is no longer TTI and \( C(t, t_w) \) depends nontrivially on both times \( t \) and \( t_w \). The meaning of aging is briefly described in the following lines. Below the glass transition temperature \( T_g \) the timescale
needed for glasses to equilibrate increases dramatically. This relaxation toward equilibrium makes some properties of the system change with time and one says that the system is aging. The effects of aging can be observed in Fig. 1.4, where the curves shift along the time axis for longer values of \( t_w \). This figure shows that the larger the value of \( t_w \) the longer the relaxation time. This means that, the closer the systems gets to equilibrium the slower it gets. The aging phenomenon has been found to behave in a very similar way in different glassy systems [13].

1.5 Dynamical Heterogeneities

A suggested possible explanation for the non–exponential relaxation in glasses (see Sec. 1.4.2) is the presence of regions within the system whose relaxation behavior substantially differs from that of other regions (Fig. 1.5). This phenomenon is called dynamical heterogeneity. One possible picture for the non-exponential relaxation is that
the relaxation process is nearly exponential in each region, but the relaxation times for different regions are different, and on average the relaxation process is non–exponential. This idea is presented in Ref. [14]. However, this is only one possibility of the relaxation process within regions and it is not a condition for dynamical heterogeneity to exist. Another believed consequence of the presence of the heterogeneous dynamics is the violation of the Stokes-Einstein relation [14–16]. This simple relation $D\eta/T = \text{const}$ between the diffusion coefficient $D$ and the viscosity $\eta$ of a liquid breaks down for supercooled liquids near the glass transition. Near this transition the relaxation time and the viscosity grows much faster than the inverse diffusion coefficient [14–16].

Dynamical heterogeneities have been observed directly both in experiments [17–23] and in simulations [16, 24]. In many instances, the particles in these regions were found to move cooperatively in clusters or strings as shown in Fig. 1.5. This figure presents two different experimental systems: a granular medium of ball bearings on the left [25] and a colloidal hard sphere suspension on the right [23].

This section is dedicated to present the different analysis used in the study of dynamical heterogeneity as well as a theoretical framework that provides a possible explanation of its origin. All quantities and concepts introduced in this section represent the major focus of my work.

1.5.1 Exponential Tails in the Particle Displacement Distributions

Exponential tails in the distribution of particle displacements have been studied in many amorphous systems [23, 26, 27]. This particular characteristic has been already established as a clear signature of dynamical heterogeneity [27].

For Fickian diffusion, the distributions are expected to follow the gaussian form

$$P_g(\Delta x, t, t_w) = \frac{1}{\sqrt{4\pi D(t - t_w)}} \exp\left[-\frac{(\Delta x)^2}{4D(t - t_w)}\right],$$

where $D$ is the diffusion coefficient and $\Delta x(t, t_w) = [x_i(t) - x_i(t_w)]$ is the displacement of a particle between time $t$
Figure 1.5: Dynamical heterogeneity observed in two different experiments. a) Granular fluid of ball bearings. Particles are color-coded by mobility, where the red particles are the most mobile ones. Reprinted by permission from Macmillan Publishers Ltd: Nature Physics (Ref. [25]), copyright (2007). b) Colloidal hard sphere suspension. The 5% most mobile particles are shown in a larger size than the rest of the particles. Colors are used to label clusters. From Ref. [23]. Reprinted with permission from AAAS.

and $t_w$. However, for glassy systems the tails of the distributions of particle displacement deviate considerably from a gaussian fit and show an exponential behavior of the form $P_e(\Delta x, t, t_w) = b \exp(-|\Delta x/x_0|^\beta)$ with $\beta$ around 1 [23, 26, 27]. It has been pointed out in Ref. [27] that the shape of the distributions can be qualitatively described by separating the particles into two groups, the “slow” particles, contribution to the center of the distribution and the “fast” particles contributing to the tails. Moreover, the tails of the distributions are known to become more pronounced close to the glass transition, when the dynamics is seen to be more heterogeneous [23, 27].

Figure 1.6 presents two examples for two different systems. The left panel of this figures shows the distribution of particle displacements of an equilibrium colloidal liquid
Figure 1.6: Exponential tails of the distribution of particle displacements for two different systems. **Left panel:** Colloidal liquid near the glass transition from Ref. [23]. Reprinted with permission from AAAS. **Right panel:** Simulation of an aging system of WCA polymers. The tails of the distribution becomes wider for larger values of $t_w$. From Ref. [26]. Copyright (2009) by the American Physical Society.

near the glass transition [23]. The right panel corresponds to a simulation of an aging system of WCA polymers [26]. The system from Ref. [26] is also used in this work in a different analysis (see Table 3.1). In both figures, the tails of the distributions are well fitted to the exponential form $P_e(\Delta x, t, t_w)$, shown as solid lines. Also, notice that for the aging system (right panel) the tails become wider with increasing waiting time $t_w$.

### 1.5.2 Dynamical Susceptibility $\chi_4$

A more quantitative information about dynamical heterogeneities can be obtained from the fluctuations of the relaxation,

$$\chi_4(t, t_w) = N \left[ \langle C(t, t_w)^2 \rangle - \langle C(t, t_w) \rangle^2 \right],$$

(1.4)

where $C(t, t_w)$ is a two–time correlation function that varies from 1 to 0, such as the ones defined in Eq. (1.2) or Eq. (1.3). The dynamical susceptibility can also be obtained from
the four-point correlation function $g_4(\vec{r}, t, t_w)$ in the following way

$$\chi_4(t, t_w) = \int d^d\vec{r} g_4(\vec{r}, t, t_w),$$

(1.5)

where

$$g_4(\vec{r}, t, t_w) = \frac{1}{N\rho} \sum_{ik} \delta(\vec{r} - \vec{r}_k(t_w) + \vec{r}_i(t_w))$$

$$\times w(\vec{r}_i(t) - \vec{r}_i(t_w))w(\vec{r}_k(t) - \vec{r}_k(t_w)) - \left(\frac{C(t, t_w)}{N}\right)^2.$$  

(1.6)

Here $\rho$ is the density of the system, and the function $w(\vec{r}_i(t) - \vec{r}_i(t_w))$ in this equation can take the form of any two-time correlation function, for instance,

$w(\vec{r}_i(t) - \vec{r}_i(t_w)) = \cos [\vec{q} \cdot (\vec{r}_i(t) - \vec{r}_i(t_w))]$ from Eq. (1.2) or

$w(\vec{r}_i(t) - \vec{r}_i(t_w)) = \theta(a - |\vec{r}_i(t) - \vec{r}_i(t_w)|)$ from Eq. (1.3). This quantity correlates the density fluctuations at two different points for two different times. Therefore, $\chi_4(t, t_w)$, the spatial integral of $g_4(\vec{r}, t, t_w)$ (see Eq.(1.5)), estimates the number of particles moving in a correlated manner.

The behavior of $\chi_4$ for an aging structural glass is shown in Fig. 1.7 (obtained from [28]). This system used to obtained this figure corresponds to a binary Lennard-Jones glass former which is also studied in this work (see Table 3.1 for further information). The peak in these curves represents the maximal correlation of the dynamics, which in this system is found to be around the timescale of $\tau_a$. As Fig. 1.7 shows, that the height of the peak of $\chi_4$ increases for longer $t_w$, i.e., when the dynamics slows down. Additionally, in the supercooled liquid regime the height of the $\chi_4$ peak increases as the system approaches the glass transition temperature [15, 24].

### 1.5.3 Four-point Structure Factor $S_4(q, t, t_w)$ and Correlation Length $\xi$

In simulations, the four-point structure factor $S_4(q, t, t_w)$ is commonly used to extract the spatial extent of dynamical heterogeneity $\xi$. This quantity is obtained by fourier
Figure 1.7: $\chi_4$ measured for a simple binary Lennard-Jones glass former. The height of the peak of $\chi_4$ is observed to increase for longer $t_w$. From Ref. [28]. Copyright (2008) by the American Physical Society.

Transforming the four-point correlation function $g_4(\vec{r}, t, t_w)$ given in Eq. (1.6). The four-point structure factor is given by

$$S_4(q, t, t_w) = N \left[ \left\langle \hat{R}(\vec{q}, t, t_w)\hat{R}(-\vec{q}, t, t_w) \right\rangle - \left\langle \hat{R}(\vec{q}, t, t_w)\right\rangle \left\langle \hat{R}(-\vec{q}, t, t_w) \right\rangle \right], \quad (1.7)$$

where

$$\hat{R}(\vec{q}, t, t_w) = \frac{1}{N} \sum_{i=1}^{N} \exp \left[ i \vec{q} \cdot \vec{r}_i(t) \right] w(\vec{r}_i(t) - \vec{r}_i(t_w)), \quad (1.8)$$

$q$ is the wave vector, the function $w(\vec{r}_i(t) - \vec{r}_i(t_w))$ is the same that was defined in Sec. 1.5.2, and $\lim_{q \to 0} S_4(q, t, t_w) = \chi_4(t, t_w)$. From Eq. (2.8) one can obtain the correlation length $\xi(t)$ by fitting it to an Ornstein Zernicke (OZ) form

$$S_4(q, t, t_w) = \frac{\chi_4(t, t_w)}{1 + [q\xi(t, t_w)]^2}. \quad (1.9)$$

The growing behavior of $\xi$ with packing fraction or temperature has been analyzed in many systems [24, 25, 29]. The correlation length increases with decreasing temperature
in structural glasses [24], or increasing packing fraction in the case of colloidal and granular systems [25, 29]. Furthermore, for aging systems the dynamical correlation length $\xi$ has been observed to increase with increasing $t_w$. This is shown in Fig. 1.8 for a binary Lennard-Jones glass former (also studied here, see Table 3.1), where the correlation length was calculated at the alpha relaxation time $\tau_\alpha$ [28].

If the correlation volume $\chi_4$ increases, the correlation length $\xi$ has to increase also, however the exact relationship between the two quantities depends on the geometry of the correlated regions, which in many cases is not known. Figure 1.9 shows the results obtained in [28] for the relationship between these two quantities at the alpha relaxation time $\tau_\alpha$. The simulated system corresponds to the same system shown in Figs. 1.7 and 1.8. The result of the fitted line $a(\xi)^b$, with $b = 2.89$ and $a = 1.23$, shown in Fig. 1.9 indicates compact clusters i.e., the obtained exponent is approximately the dimension of the system.

Figure 1.8: The dynamical correlation length as a function of waiting time $t_w$, measured for a simple binary Lennard-Jones glass former. The correlation length was obtained at the alpha relaxation time, i.e., when $C(t, t_w) = 1/e$. From Ref. [28]. Copyright (2008) by the American Physical Society.
Figure 1.9: The dynamical susceptibility versus the correlation length for different waiting time $t_w$, measured for a simple binary Lennard-Jones glass former. Both quantities were obtained at $\tau_\alpha$. The solid line corresponds to the fit $a(\xi_4)^b$, with $b = 2.89$ and $a = 1.23$. From Ref. [28]. Copyright (2008) by the American Physical Society.

However, the shape of the clusters may depend on the timescale: in the case of hard spheres in 3D, it has been found that the exponent goes from being close to 3 for $\Delta t \approx \tau_\alpha$ to being around 2 for $\Delta t >> \tau_\alpha$ [29].

1.5.4 Time Reparametrization Invariance

The dynamics of some spin–glass models in the aging regime has been proven to be invariant under time reparametrization, $t \rightarrow h(t)$ [30–33], in the limit of very long times. This invariance indicates that there is a family of continuous symmetries for those systems, which is broken by dynamical correlations and responses close to the glass transition. For instance, if one applies a transformation $t \rightarrow h(t) = 2t$ to the correlation function $C(t, t_w)$ we obtain $\hat{C}(t, t_w) = C(h(t), h(t_w)) = C(2t, 2t_w)$. To be more specific, let me assume that $C(t, t_w)$ is described by Eq. 1.1, therefore such transformation leads to
exp \{-2(t - t_w)/\tau\} \neq \exp \{-[(t - t_w)/\tau]\}, \text{ i.e., } \hat{C}(t, t_w) \neq C(t, t_w), \text{ and, in this case, the relaxation time is reduced: } \tau \rightarrow \tau' = \tau/2. \text{ This means that if we only consider the dynamics at timescales much longer than typical vibrational periods the time reparametrization symmetry is broken by } C(t, t_w). \text{ In contrast, in the case of liquids above the melting temperature, the relaxation time is very short, so at time differences } t - t_w \text{ much longer than typical vibrational timescales, } C(t, t_w) = 0. \text{ If we apply an arbitrary reparametrization, as long as we stay within timescales much longer than typical vibrational periods, we get } \hat{C}(t, t_w) = C(h(t), h(t_w)) = 0 = C(t, t_w), \text{ i.e. the symmetry is not broken. In summary, the symmetry is broken only when the relaxation is very slow, namely in the supercooled liquid and in the glass state.}

As with any broken continuous symmetry, we expect collective modes to appear. These collective modes are called Goldstone modes \[34\]. For instance, in the ground state of a lattice of isotropic ferromagnetically coupled Heisenberg spins all spins are parallel (breaking their continuous rotational symmetry), but little cost in energy is paid by slowly varying the direction of the spins over the sample. In Refs. \[35\] and \[36\], a Goldstone mode associated with non–uniform time reparametrizations was postulated to explain the behavior of fluctuations in the aging regime for a model of spin glasses.

This hypothesis states that dynamical heterogeneities emerge from fluctuations modeled as Goldstone modes which are associated with spatially varying time reparametrizations, \( t \rightarrow h_\vec{r}(t), \text{ i.e.} \)

\[ C_\vec{r}(t, t_w) = C(h_\vec{r}(t), h_\vec{r}(t_w)). \] \hspace{1cm} (1.10)

\footnote{This form of the hypothesis is actually too restrictive. In principle, the very presence of the fluctuations will make the \( C(t, t_w) \) function appearing in the rhs of Eq (1.10) different from the measured global two–time correlation. We ignore this difference in all of the work discussed here, which has the effect of making the time reparametrization fluctuations seem to be less dominant than they actually are.}
Here, $C(t, t_w)$ is a global\(^2\) two-time correlation function like the one defined in Eq. (1.2) or Eq. (1.3). The corresponding local two-time correlation function [37] for the case of Eq. (1.2) is given by

$$C_r(t, t_w) = \frac{1}{N(B_r)} \sum_{\vec{r}(t_w) \in B_r} \cos(q \cdot (\vec{r}(t) - \vec{r}(t_w))), \quad (1.11)$$

where $B_r$ denotes a small coarse-graining box around the point $\vec{r}$, and the sum runs over the $N(B_r)$ particles present in the box at the waiting time $t_w$. The aforementioned hypothesis implies that different regions in the systems are relaxing at different rates, i.e. the dynamics is heterogeneous. However, Eq. (1.10) implies that these fluctuations should be all concentrated along a trajectory corresponding to the evolution of global quantities describing the system [35, 36]. This idea is shown in the schematic Fig. 1.10 where three different regions $x_A$, $x_B$ and $x_C$ are following the same relaxation path (green solid arrow), but the relaxation of region $x_A$ is advanced (retarded), in the time axis, with respect to region $x_B$ and $x_C$ if one concentrates in the solid red line (dashed red line).

Indirect evidence of the presence of Goldstone modes for structural glasses was presented in Ref. [37]. This test is shown in Fig. 1.11. The results show that when the global correlation function $C(t, t_w)$ is kept constant, the probability distributions for the local fluctuating two-time quantities, $\rho(C_r(t, t_w))$ and $\rho(\Delta x(t, t_w))$, are approximately independent of $t_w$. In principle, the approximate scaling of the distributions obtained from local quantities, when $C(t, t_w)$ is kept constant, can be associated with the presence of the time reparametrization symmetry with the local fluctuations related to the presence of Goldstone modes as described in Eq. (1.10). However, a more direct test is required to attribute these local fluctuations to the presence of Goldstone modes in structural glasses. A better approach would be a direct test of Eq. (1.10).

\(^2\) From here on the term “global” will be used to describe quantities evaluated over the whole system, as opposed to “local” quantities which are evaluated over a small region of the system.
Figure 1.10: Illustration of fluctuations in the time variable. The green curve represents the time evolution of the regions, with the red marks indicating the age of each region (Figure courtesy of H. Castillo).

1.5.4.1 Triangular Relations

A more direct test than the one described in Fig. 1.11 was presented in Ref. [33] for a spin glass system. This method represents part of the inspiration for the work presented in Chapter 3.

To introduce this method first allow me to explain the motivation. It would be desirable to define a trajectory for the global quantities that would describe the temporal evolution of the system, like the one shown in the illustration in Fig. 1.10. If we construct a set of parametric plots of $C(t_2, t_1)$ against $C(t_3, t_2)$ using $t_2$ as a parameter varying between $t_3$ and $t_1$, with $t_1 < t_2 < t_3$, we will be moving along one of the curves shown in Fig. 1.12. If the time $t_2$ is closer to $t_1$, the point $(C(t_3, t_2), C(t_2, t_1))$ is located in the upper left side of the curve. As $t_2$ is incremented closer to $t_3$, the position of the point will move towards the bottom right part of the curve. In this sense, the position of the point in the
Figure 1.11: Probability distributions for the local fluctuation of a two–time correlation function. Reprinted by permission from Macmillan Publishers Ltd: Nature Physics (Ref. [37]), copyright (2007). **Left panel:** Probability distributions $\rho(C_r(t, t_w))$ for values of $t_w$ in the range [30.20:30200]. **Right panel:** Probability distributions $\rho(\Delta x(t, t_w))$ for values of $t_w$ in the range [30.20:30200]. In both panels the distributions are plotted for final times $t$ chosen in the way that $C(t, t_w) = 0.1, 0.3, 0.5,$ and 0.7.

A parametric curve describes the evolution of the relaxation process between the time $t_1$ and $t_3$. However, different values of $(t_1, t_3)$ give different curves (see Fig. 1.12). Since one would like to make a comparison of the local fluctuations at all times, this shifting of the curve is undesirable. In the following paragraphs I present a way to overcome this barrier presented also in [33].

For a spin glass system, it has been found that the function $f$ for the slow part of the relaxation\(^3\) $C(t_1, t_3) < q_{EA}$ is compatible with the scaling form [33]:

$$C(t, t_w) = f\left(\frac{h(t)}{h(t_w)}\right).$$

\(^3\) Analogous to the $\alpha$ relaxation in a structural glass, see Sec. 1.4.2.
The assumption that the correlation is a monotonic function allows the derivation of some of its properties. In principle, if three long times are chosen so that \( t_1 < t_2 < t_3 \), the correlation \( C(t_3, t_1) \) can be broken down as follows [33, 38]:

\[
C(t_3, t_1) = f \left( \frac{h(t_3)}{h(t_1)} \right) = f \left( \frac{h(t_2)}{h(t_1)} \frac{h(t_3)}{h(t_2)} \right) = f \left( f^{-1} \left[ C(t_2, t_1) \right] f^{-1} \left[ C(t_3, t_2) \right] \right).
\] (1.13)

This last equation can be also written as

\[
f^{-1} \left[ C(t_3, t_1) \right] = f^{-1} \left[ C(t_2, t_1) \right] f^{-1} \left[ C(t_3, t_2) \right].
\] (1.14)

If we define \( \Psi_{ab} = f^{-1} \left[ C(t_a, t_b) \right] \), with \( a, b, \in \{1, 2, 3\} \), then, we obtain the triangular relation

\[
\Psi_{31} = \Psi_{21} \Psi_{32}.
\] (1.15)

Figure 1.12: Parametric plot of the global correlation functions \( C(t_1, t_2) \) against \( C(t_2, t_3) \) using \( t_2 \) as a parameter, by taking values between \( t_1 \) and \( t_3 \), for four different values of \( t_3 \). Data from the simulation of a system of WCA polymers (See Table 3.1).
This suggests the choices $X \equiv \Psi_{32}/\sqrt{\Psi_{31}}$ and $Y \equiv \Psi_{21}/\sqrt{\Psi_{31}}$. Therefore, if we know $f$, then the parametric plot for different values of $t_3$ and $t_1$ can be made to collapse to a single curve $Y = 1/X$, as represented in Fig. 3.2.1.2.

If time reparametrization invariance is present, the coarse-grained local correlation should scale in a similar way as the global correlation [35, 36], i.e.,

$$C_r(t, t_w) \approx f\left(\frac{h_r(t)}{h_r(t_w)}\right).$$  \tag{1.16}

Again, this equation implies that any three local correlations between three times $t_1$, $t_2$ and $t_3$, where $t_1 < t_2 < t_3$, are related by [33]

$$C_r(t_3, t_1) \approx f\left(f^{-1}[C_r(t_2, t_1)] f^{-1}[C_r(t_3, t_2)]\right).$$  \tag{1.17}

In other words, they satisfy the same triangular relation as the global quantities, and therefore should collapse on top of the global curves, i.e., $1 = X_r Y_r$. However, they do not collapse to a point because the dynamics is heterogeneous, i.e., the parametric construction for the local correlations should have some regions with the local value of $h_r(t_2)$ being younger or older with respect to the global value, which puts them at different locations along the global curve. Other kind of fluctuations could make the points separate in the direction perpendicular to $1 = XY$. However, the time reparametrization fluctuations are expected to be stronger and therefore most of the points should be located near the global curve, as illustrated in Fig. 1.13. A related way of testing this prediction is that if we plot the 2d projection of the joint probability density of the coarse-grained correlations, the contours of constant probability density should all extend along the global curve and not away from it. This prediction has been tested for a spin glass system in Ref. [33]. For the case of structural glasses, a less robust version of the triangular relation was presented in [39]. In this first test, the quantities on the axes had the forms $X \equiv C(t_3, t_2)/C(t_3, t_1)^\alpha$ and $Y \equiv C(t_2, t_1)/C(t_3, t_1)^\alpha$. The value of $\alpha$ was dependent on the system and it was found by trial and error. This resulted in a not well collapsed global
curve. Nevertheless, the anisotropy of the 2d contours of the joint probability density \( \rho(X, Y) \) was observed to be extended in the direction of the global curve. Even when this test gave a good indication of the presence of Goldstone modes in structural glasses, in principle, a more complete analysis could be done by finding the functional form of \( f(x) \). Moreover, the analysis in [39] only focuses on temperatures below the mode coupling critical temperature \( T_c \) [3]. In principle, one can apply this test to the same systems at higher temperatures \( T > T_c \), when the time reparametrization fluctuations are expected to be weak, and compare the results to the results obtained at \( T < T_c \).

1.6 Organization of the Dissertation

In Chapter 2, I present a study of dynamical heterogeneity in a dissipative granular fluid. Here, the study focuses in the analysis of quantities typically used to study dynamical heterogeneity in structural glasses but which have been up to now poorly
explored for granular fluids. Some of these quantities were introduced in Sec. 1.5. Mainly, the goal of this project is to provide a better understanding of the way dynamical heterogeneity can be compared in granular materials and in other glass formers. Also, since this is a dissipative model, one can compare results with those for systems of non-dissipative hard spheres.

In Chapter 3, I use numerical simulation data for structural glasses to test the theoretical framework presented in Sec. 1.5.4. Beside using the triangular relations (see Sec. 1.5.4.1) to test the hypothesis, this work extends to a more quantitative analysis of the fluctuations related to time reparametrizations and a comparison to other fluctuations in the system.

Altogether, this work addresses two basic question in the field: how the dynamical regions in glass formers are comparable to those of a granular system? and why do we have dynamical heterogeneity?

Finally, in Chapter 4, I discuss general conclusions of this work and some possible future directions for further research.
2 Project 1: Dynamical Heterogeneity in a Dissipative Granular Fluid

In this chapter, I present the results of a study of dynamical heterogeneity in a dissipative granular fluid. This work was done in a collaboration with the research group of Prof. Annette Zippelius at the University of Göttingen, Germany. Two manuscripts are being prepared from the results presented in this chapter. The details of the model and the simulation are described in Sec. 2.1. As mentioned before, because granular media are more easily accessible to experiments that track the microscopic configurations of the particles, it is desirable to establish a relationship between them and other glassy systems. Moreover, the fact that the system studied here is based on a dissipative model gives the opportunity to make comparisons with other simulations of hard spheres with fully elastic collisions. In addition, the dissipative nature of the collisions imposed in this system makes it more comparable to typical experiments in granular fluids.

In general, this project focuses on dynamical heterogeneities by studying the growing behavior of the dynamical susceptibility $\chi_4(t)$ and the extent of the dynamical heterogeneity $\xi(t)$ as a function of packing fraction $\phi$. Here, the correlation length $\xi(t)$ is obtained by calculating the Fourier-transformed four-point correlation function $S_4(q, t)$ (see Sec. 1.5.3) via the overlap correlation function. The results of the analysis of dynamical heterogeneity as a function of packing fraction are introduced in a qualitative way in Sec. 2.2.1 and then in a more quantitative way in Sec. 2.2.2. Furthermore, the dependence of $\xi$ and $\chi_4$ on the cutoff length $a$ selected in the overlap function is explored in detail for the first time in Sec. 2.2.3. It is known that $\chi_4(t)$ can have a strong dependence on $a$, but surprisingly, there are few studies that focus on this subject [41–44]. Another interesting question in dynamical heterogeneity is the lifetime of the correlated regions. For this system, this question is addressed in Sec. 2.2.4. Finally, the dependence
on the strength of the dissipation in the particle collisions is analyzed in Sec. 2.2.5. Since this effect can substantially affect the properties of the fluctuations, a better understanding of the effect of the energy dissipation would be particularly desirable. At the end of this chapter, I give a short summary and discuss all the results obtained in this project.

2.1 Model and Simulation Details

The model consists of a 2D system of hard disks which only interact via two-body inelastic collisions, without rotational degrees of freedom. The system is bidisperse, i.e., it is composed of particles of two sizes, with concentration ratio 1 : 1. In this work, the ratio of particles radii is given by \( r_2/r_1 \approx 1.43 \), where \( r_1 \) denotes the radius of the small particles and \( r_2 \) denotes the radius of the large particles. The change in the velocities of two colliding particles, particle \( i \) and particle \( j \), is given by

\[
(\vec{g} \cdot \vec{n})' = -\varepsilon (\vec{g} \cdot \vec{n}),
\]

(2.1)

where \( \vec{g} = \vec{v}_i - \vec{v}_j \) is the relative velocity, \( \vec{n} = (\vec{r}_i - \vec{r}_j)/|\vec{r}_i - \vec{r}_j| \) is a unit vector that connects the center of the two disks and \( \varepsilon \) corresponds to the coefficient of restitution, which is a constant (\( \varepsilon = 1 \) in the elastic case). The primed quantities refer to post-collisional velocities while the unprimed ones refer to pre-collisional velocities. Therefore, the velocities of the two disks after a collision are given by

\[
m_i\vec{v}_i' = m_i\vec{v}_i - m_im_j\frac{(1 + \varepsilon)}{2}(\vec{g} \cdot \vec{n})\vec{n},
\]

(2.2)

and

\[
m_j\vec{v}_j' = m_j\vec{v}_j + m_im_j\frac{(1 + \varepsilon)}{2}(\vec{g} \cdot \vec{n})\vec{n},
\]

(2.3)

where \( m_i \) corresponds to the mass of particle \( i \). Here, constant mass density is assumed for all particles such that the mass ratio of the particles is given by \( (r_2/r_1)^2 \).

In these systems, the driving of the particles is important to compensate the energy dissipation due to collisions. In experiments, this driving is usually done by shearing,
shaking, air tables or similar methods. Here, the energy is fed homogeneously by bulk driving. This is comparable to the bulk driving in the experiments presented in [25]. This method for feeding energy consists of applying an instantaneous kick to a given particle at time \( t \)

\[
\vec{v}_i'(t) = \vec{v}_i(t) + v_{Dr} \vec{k}_i(t),
\]

(2.4)

where \( v_{Dr} \) is the constant driving amplitude and \( \vec{k}_i(t) \) is a random direction. To ensure momentum conservation, pairs of particles are kicked in opposite directions [45].

This is the same system as the one presented in Ref. [46]. This reference can also be consulted for additional details of the simulation. In this work, I analyze data from simulations carried out by Dr. Andrea Fiege on systems containing \( N_{\text{tot}} = 4,000,000 \) particles for the packing fractions \( \phi = 0.60, 0.65, 0.70, 0.72, 0.74, 0.76 \) and \( 0.78 \), which were also used in Ref. [46] in a different analysis. Also, I analyze simulation data obtained by Dr. Iraj Gholami and me for \( N_{\text{tot}} = 360,000 \) for packing fractions \( \phi = 0.805, 0.80 \) and \( 0.79 \). All simulations mentioned above correspond to the coefficient of restitution \( \varepsilon = 0.90 \). Simulations for different values of the coefficient of restitution \( \varepsilon = 1.00, 0.80 \) and \( 0.70 \) were performed by Dr. Andrea Fiege and me for \( \phi = 0.79, 0.78, 0.76 \). Moreover, for each packing fraction \( \phi \) I selected the time window such that the system was in the steady state.

The results shown in this chapter are presented in reduced units, where the length units correspond to \( r_1 \) and the mass units correspond to \( m_1 \). Also, the time steps units are set such that \( \frac{1}{2}m_i\vec{v}_1^2(t = 0) = 1 \) (averaged over all particles).

**2.1.1 Averages in our Results**

One important aspect in the analysis of the granular data is the way the calculated quantities are averaged. Before introducing the results, this section is dedicated to explaining the main procedures used to average data in this chapter, namely averaging
over space and averaging over time. Also, the notation used in the equations that indicates a specific type of average is established here.

2.1.1.1 Space Average

The notation \( \langle \cdots \rangle \) indicates when space average is performed in any calculation. In order to perform space averages in the analysis, the simulated box of total area \((L_{\text{tot}})^2\), which contains the \(N_{\text{tot}}\) particles, was divided into sub-boxes of equal area \(L^2\), as shown in the schematic Fig. 2.1. In principle, one can select the length \(L\) such that the sub–boxes accommodate, on average, a desired number of particles \(N\). This can be important depending on the analysis performed. The choice of \(N\) will be specified in each result. However, even when \(N\) is fixed, the number of particles per sub–box, \(N_{Br}\), and the particle concentration can vary between different sub–boxes. The notation \(N_{Br}\) denotes the number of particles of a sub–box centered on a point \(\vec{r}\).

Notice that the total number of sub–boxes can be calculated by the ratio \(N_{\text{tot}}/N\). In general, the more particles \(N_{\text{tot}}\), the better the statistics in the analysis.

Figure 2.1: The simulation box of total area \((L_{\text{tot}})^2\), which contains \(N_{\text{tot}}\) particles, is divided into sub–boxes of area \(L^2\), which contain a number \(N_{Br}\) of particles that might vary between sub–boxes. Please note that the sub–boxes are allowed to overlap since the origin of each one is arbitrary.
2.1.1.2 Time Average

The notation \( \cdots \) in the equations specifies that time average is performed. Besides space averaging, I use time averaging for some calculations to improve the statistics, especially for the packing fractions \( \phi = 0.79, 0.80 \) and 0.805, for which the system contains fewer particles than for the rest of the packing fractions. It is worth noting that time averaging is possible because the simulations are done in the stationary state, i.e., the system is not aging. This means that an average over different choices of \( t_w \) can be performed for quantities that have a dependence on the time difference, \( \Delta t = t - t_w \).

2.2 Results

Most of the quantities used in the following analysis are defined in Chapter 1. However, to allow for the possibility of aging, the quantities defined in Chapter 1 can in principle depend on the waiting time \( t_w \). Since this granular fluid is in the stationary state, TTI is recovered and therefore, there is only dependence on the time difference \( \Delta t \) and not on the waiting time \( t_w \). To avoid confusion, all quantities are redefined in this section by dropping the notation \( t_w \) and using \( t_0 \) instead.

2.2.1 Particle Displacements

To visually observe dynamical heterogeneity in this system the particles can be classify by color–coding according to their mobility. This idea has been used widely for other systems (see for example Fig. 1.5). Here, the mobility of each particle at time difference \( t \) is defined by \( (r_i(t_0 + t) - r_i(t_0))^2 \). The result of applying this method is presented in Fig. 2.2. This figure shows three different packing fractions at their corresponding relaxation time \( \tau_\alpha \). The 10% fastest particles are displayed in gray and the 10% slowest particles in red.
Figure 2.2: Particles are color-coded by mobility, where gray particles are the 10% slowest ones and the red particles are the 10% fastest ones for different packing fractions: (a) $\phi = 0.60$, (b) $\phi = 0.78$ and (c) $\phi = 0.805$. The correlation length grows as the system gets closer to the jamming transition.

It can be observed that subsets of particles moving at similar rates form clusters or strings for high packing fractions. Moreover, this figure shows that the size of the clusters and strings increases for densities closer to the jamming transition, which for this granular system was determined to be at $\phi_j \approx 0.818$ (see Sec. 2.2.2).

To estimate the mobility of the slow and fast clusters one can look at the mean square displacement (MSD) calculated only for these different subsets of particles. The MSD calculated from all particles is given by

$$\Delta(t) = \frac{1}{N_{\text{tot}}} \sum_{i=1}^{N_{\text{tot}}} (\vec{r}(t_0 + t) - \vec{r}(t_0))^2,$$

where $N_{\text{tot}}$ is the total number of particles in the simulated box (see Sec. 2.1.1 for the definition of $N_{\text{tot}}$) and $\vec{r}(t)$ is the position of particle $i$ at time $t$. Notice that in order to calculate the MSD for subsets of particles, the number of particles in Eq. (2.5) should be modified accordingly. Fig. 2.3 shows the MSD for different subsets of particles as a
function of time for the packing fraction $\phi = 0.78$. The MSD was estimated for certain percentages of fast and slow particles extracted either from all particles (solid lines) or only from small particles (dotted–dashed lines). For comparison, the MSD calculated using all particles and all small particles is also displayed in this figure. We can observe that while fast particles are moving during the entire simulation several times the radius of the small particles $r_1$, slow particles are barely moving. The drastic differences of cluster mobility gives us an idea of the strength of the dynamical heterogeneity.

As discussed in Sec. 1.5.1, exponential tails in the particle displacement distributions have been already established as a clear signature of dynamical heterogeneity and have
been observed in different glassy systems. For this granular fluid, the calculated
distributions of small particle displacements can be observed in Fig. 2.4. The top panel of
this figure corresponds to the distributions for the packing fractions $\phi = 0.805$, 0.78, and

Figure 2.4: **Top panel:** Distribution of small particle displacements for different packing
fractions $\phi = 0.805$, 0.78, and 0.60 (from right to left) at time $\tau_\alpha$. **Bottom panel:**
Distribution of small particle displacements for $\phi = 0.78$ at different times $t = 0.01\tau_\alpha$,
$t = \tau_\alpha$ and $t = 8.5\tau_\alpha$ (from left to right). The tails of the distributions are better described
by an exponential fit (solid lines) than by a gaussian fit (dotted-dashed lines).
0.60 at time $\tau_\alpha$. The bottom panel corresponds to the distributions at different times for the intermediate packing fraction shown in the top panel, $\phi = 0.78$. The corresponding times shown in this figure are $t = 0.01 \tau_\alpha$, $t = \tau_\alpha$ and $t = 8.5 \tau_\alpha$ (from left to right). The gaussian fit to the data, $P_g(\Delta x, t) = (1/4\pi Dt) \exp[-(\Delta x)^2/(4Dt)]$, is also shown in Fig. 2.4 by the dotted-dashed lines, where $D$ was set as a fitting parameter. We can observe that the tails of the distributions for all packing fractions and times deviate considerably from the gaussian fit and follow an exponential behavior of the form $P_e(\Delta x, t) = b \exp(-|x/x_0|)$, shown as solid lines in Fig. 2.4. These results are qualitatively similar to the results seen in glasses (see Sec. 1.5.1). Also, notice that the tails become wider when increasing packing fraction and time.

### 2.2.2 Dynamical Susceptibility $\chi_4$ and Correlation Length $\xi$ as Functions of $\phi$

The most typical quantities used in glassy systems to characterize the dynamics are two-time correlation functions, which capture the fraction of slow particles at a given time interval (see Sec. 1.4.2). Here, the overlap function is used to analyze the granular data. The overlap function is given by

$$C_a(t) = \frac{1}{N_{Br}} \sum_{i=1}^{N_{Br}} \theta(a - |\vec{r}_i(t_0 + t) - \vec{r}_i(t_0)|),$$

(2.6)

where $N_{Br}$ is the number of particles in a sub-box centered at a point $\vec{r}$ and $\theta = 1$ if $|\vec{r}_i(t_0 + t) - \vec{r}_i(t_0)| \leq a$, or zero otherwise. The following results are calculated for $a = 0.6 r_1$ and the coefficient of restitution $\varepsilon = 0.9$, unless indicated otherwise. Different choices of $a$ and $\varepsilon$ are discussed in Sec. 2.2.3 and Sec. 2.2.5, respectively. Fig. 2.5 shows $\langle C_a(t) \rangle$ for different packing fractions. It can be clearly seen that the time scale increases with increasing packing fraction $\phi$, which is the expected result as the dynamics becomes more sluggish when increasing packing fractions. The increase of the alpha relaxation time $\tau_\alpha$ with packing fraction can be better observed in Fig. 2.6. The curve is fitted to an inverse power law $\tau_\alpha \propto (\phi_J - \phi)^{-\gamma_r}$ (dashed line) as predicted by mode coupling theory [3], and to
Figure 2.5: Overlap function, \( < C_a(t) > \), for different packing fractions, \( \phi = 0.60, 0.65, 0.70, 0.72, 0.74, 0.76, 0.78, 0.79, 0.80 \) and 0.805. The time scale increases with increasing \( \phi \). The black horizontal line corresponds to the value of the correlation function at which \( \tau_\alpha \) is defined i.e., \( < C_a(\tau_\alpha) > = 1/e \).

The number of particles moving together in a correlated manner is given by the dynamical susceptibility

\[
\chi_4(t) = N[\langle C_a(t) \rangle^2 - \langle C_a(t) \rangle^2],
\]

(2.7)

where \( N \) corresponds to the average number of particles over all sub-boxes. This equation quantifies the fluctuations of the overlap correlation function given by \( C_a(t) \). In Fig. 2.7,
we can observe that the peak value of $\chi_4^P(t)$, $\chi_4^P$, increases with increasing $\phi$. This behavior is an indication of a growing correlation length, $\xi$, closer to the jamming transition.

Moreover, as shown in Fig. 2.8, the peak of the dynamical susceptibility, $\chi_4^P$, increases as a function of system size and then saturates. Both the value of $\chi_4^P$ and the value of the system size at which $\chi_4^P$ reaches a plateau increase with increasing $\phi$. This finite-size scaling behavior also suggests the existence of a growing dynamical correlation length. A more detailed finite-size scaling analysis was presented for a simulated glass-former by Karmakar et al. in Ref. [49].
Figure 2.7: Dynamical susceptibility, $\chi_4(t)$, for packing fractions $\phi = 0.60, 0.65, 0.70, 0.72, 0.74, 0.76, 0.78, 0.79, 0.80$ and $0.805$. The peak value of $\chi_4(t)$ increases with $\phi$, which suggests an increasing correlation length.

As mentioned in Sec. 1.5.3, the four-point structure factor $S_4(q, t)$ is widely used in simulations to quantitatively obtain the value of the correlation length. $S_4(q, t)$ is given by

$$ S_4(q, t) = \left\{ N \left[ \langle \hat{R}(q, t)\hat{R}(-q, t) \rangle - \langle \hat{R}(q, t) \rangle \langle \hat{R}(-q, t) \rangle \right] \right\}, \quad (2.8) $$

where

$$ \hat{R}(q, t) = \frac{1}{N} \sum_{i=1}^{N} \exp \left[ i \mathbf{q} \cdot \mathbf{r}_i(t_0) \right] \theta(a - |\mathbf{r}_i(t_0) + t - \mathbf{r}_i(t_0)|), \quad (2.9) $$

and $\langle \cdots \rangle$ denotes an average over wave vectors $\mathbf{q}$ of equal magnitude. The results obtained for $S_4(q, \tau_\alpha)$ are shown in the top panel of Fig. 2.9. The correlation length $\xi(\tau_\alpha)$ can be obtained from the results of $S_4(q, \tau_\alpha)$ by an Ornstein-Zernike (OZ) fit,

$$ S_4(q, \tau_\alpha) = \frac{\chi_4(\tau_\alpha)}{1 + [q\xi(\tau_\alpha)]^2}. \quad (2.10) $$

The correlation length for different packing fractions obtained from the OZ fit are written in the key label of the bottom panel of Fig. 2.9. These results confirm the observations
Figure 2.8: Peak value of the dynamical susceptibility, $\chi^P_4$, versus $N$ for the packing fractions $\phi = 0.60, 0.65, 0.70, 0.72, 0.74, 0.76, 0.78, 0.79, 0.80$ and $0.805$. $\chi^P_4$ grows with $N$ before it saturates and it also grows as $\phi$ is increased.

from the previous section and the observation for the growing peak of $\chi_4(t)$. The extent of dynamical heterogeneity does grow rapidly with packing fraction. The values of $\xi(\tau_a)$ found in this system at high values of $\phi$ are much larger than the values found for other granular systems (glasses) close to the jamming (glass) transition [15, 24, 29, 41, 49]. The quality of the value of $\xi(\tau_a)$ obtained from the OZ fit can be determined by the scaling plot $S_4(q, \tau_a)/S_4(q \to 0, \tau_a)$ against $q\xi(\tau_a)$. It can be observed in this figure that the curves follow the fit for values $q\xi(\tau_a) \lesssim 3$. This is a good estimation since the important dynamics is concentrated at small values of the wave vector $q$. In addition to the aforementioned point, there is a good collapse between the data at different packing fractions. The fitting range used to estimate the values of $\xi(\tau_a)$ shown in Fig. 2.9 is $[0 : 0.2]$ for all packing fractions. To minimize finite size effects, the values of $N$ used in the analysis of $S_4(q, \tau_a)$ were chosen such that the value of $\chi^P_4$ had saturated, i.e., at the
Figure 2.9: Top panel: Four–point structure factor $S_4(q, \tau_\alpha)$ for different packing fractions. The values at $q = 0$ were obtained by the direct calculation of Eq. (2.8). Bottom panel: Scaling plot of the four–point structure factor $S_4(q, \tau_\alpha)$ for different packing fractions. The solid line corresponds to the OZ fit. As shown in the key, the correlation length $\xi(\tau_\alpha)$ grows rapidly with growing $\phi$.

Plateau of Fig. 2.8. From this point on, all results are calculated for $N = 10,000$ for $\phi \leq 0.76$ and $N = 40,000$ for $\phi > 0.76$. 
In addition to fitting $S_4(q, \tau_\alpha)$ to an OZ function, $S_4(q, \tau_\alpha)$ was fitted to different forms:

$$S_4(q, \tau_\alpha) = \frac{\chi_0}{1 + (q\xi)^2 + A^2(q\xi)^4} \frac{B}{[1 + (q\xi)^2]^2},$$

(2.11)

suggested in [29, 50]. The left panel of Fig. 2.10 shows the results of $\xi(\tau_\alpha)$ obtained by using different maximum values of $q$ in the fitting range, $q_m$, for the following fitting forms: Fit (1) corresponds to the set of parameters $\chi_0 = \chi_4(t)$, $A = 0$ and $B = 0$, which is the OZ form in Eq. (2.10); Fit (2) corresponds to $A = 0$ and $B = 0$; Fit (3) corresponds to $B = 0$; and Fit (4) corresponds to the set of parameters $\chi_0 = \chi_4(t)$ and $A = 0$. This figure only show results for the packing fractions $\phi = 0.805, 0.80, 0.79$ and 0.78. The same analysis was performed for the other packing fractions, which for the sake of clarity is not shown in Fig. 2.10. It was found that, specially for $\phi \leq 0.78$, different fitting functions did not change substantially our results. This holds for the whole range of $q_m$ tested. It was also found that the values of $\xi(\tau_\alpha)$ obtained from the different fits do not depend strongly on the fitting range $[0, q_m]$. This is true except for very small values of $q_m$, in which case the deviations could be attributed to the small number of data points. Therefore, further calculations of $\xi$ will be obtained similarly to those obtained in Fig. 2.9, i.e., using the OZ form to fit $S_4(q, t)$ up to $q_m = 0.2$.

It is important to remark that, as demonstrated in Ref. [51], for numerical simulations where the particle density and the relative concentrations of each particle type are held fixed, additional contributions to the right hand side of Eq. (2.7) are expected [29, 52]. In this work, since the simulated box is cut into sub–boxes of equal size, the number of particles and particle concentration varies between sub–boxes. Therefore, all fluctuations are expected to be accounted for in Eq. (2.7) and no additional terms are needed in the calculation of $\chi_4(t)$. This means that the $q \rightarrow 0$ limit of $S_4(q, t)$ is expected to be well described by $\chi_4(t)$ obtained from the calculation of Eq. (2.7), whereas for other ensembles,
Figure 2.10: Left panel: $\xi(\tau_\alpha)$ dependence on the fitting range $[0, q_m]$ of four fitting functions (described in the text) shown for $\phi = 0.78, 0.79, 0.80$ and 0.805. The values of $\xi(\tau_\alpha)$ for a given packing fraction are fairly similar within all $q_m$ compared and also $\xi(\tau_\alpha)$ between different fitting forms are very similar for all values of $q_m$. Right panel: Comparison of $\chi_4(\tau_\alpha)$ obtained from Eq. (2.7), shown by the dotted–dashed line, to $\chi_0$ obtained by fitting functions 2 and 3, shown as symbols. The values of $\chi_0$ obtained from both fitting functions are very close to $\chi_4$, even for large $q_m$.

$S_4(q = 0, t)$ is obtained by an extrapolation of $\lim_{q \to 0} S_4(q, t)$ [24, 29] or by directly calculating the missing contribution to $\chi_4(t)$ as presented in Ref. [29]

To confirm whether Eq. (2.7) gives a reliable value of $S_4(q = 0, \tau_\alpha)$ the values of $\chi_0$ obtained from fits (2) and (3), shown as symbols in the right panel of Fig. 2.10, are compared with the value of $\chi_4(\tau_\alpha)$ obtained from Eq. (2.7), shown as the dotted-dashed line in the same figure. The agreement between $\chi_0(t)$ and $\chi_4(t)$ is remarkably good even for packing fraction with less statistics, $\phi \geq 0.79$.

The trend of $\xi(\tau_\alpha)$ and $\chi_4(\tau_\alpha)$ as functions of $\phi$ can be observed in the top panel of Fig. 2.11. In this figure, $\chi_4(\tau_\alpha)$ against $\phi$ is fitted to an inverse power law
$\chi_4(\tau_a) \propto (\phi_J - \phi)^{-\gamma_\chi}$ predicted by mode coupling theory [3]. Similarly, in the same figure, the values of $\xi(\tau_a)$ obtained from Fig. 2.9 against $\phi$ are fitted to $\xi(\tau_a) \propto (\phi_J - \phi)^{-\gamma_\xi}$. In both cases, $\phi_J$ was fixed to the value 0.818 obtained in Fig. 2.6. The values of the exponents were found to be $\gamma_\chi = 2.04$ and $\gamma_\xi = 1.32$.

As already mentioned $\chi_4(t)$ measures the number of particles moving together in a cooperative manner, and $\xi(t)$ is the approximate extension of these cooperative regions in one direction. Thus, from the relationship $\chi_4(t) \approx \mathcal{A}\xi(t)^{d-p}$ the exponent $d - p$ is usually interpreted as the fractal dimension of the clusters [15]. For instance, this would mean that the case of $d - p$ equal to the dimensionality $d$ of the system corresponds to compact clusters, whereas the case of $d - p = 1$ corresponds to strings. The scaling behavior of $\chi_4(\tau_a)$ versus $\xi(\tau_a)$ for this system is shown in the bottom panel of Fig. 2.11, with the fitted value $d - p \approx 1.63$.

An additional desired analysis with the quantities already shown would be the relationship between time scales and length scales. In Figs. 2.6 and 2.11 it was determined that $\tau_a$ and $\xi(\tau_a)$, respectively, grow as the jamming transitions approaches. Next, the relation between them is explored in Fig. 2.12. In this figure, the points are fitted to a power law $\tau_a \propto \xi(\tau_a)^z$, with $z = 1.47$ (solid line), predicted by the mode coupling theory, and to an exponential form $\tau_a \propto \exp[k\xi(\tau_a)]$ (dotted-dashed line). It was found that, in the whole fitting range, the power law function describes better the relationship between $\tau_a$ and $\xi(\tau_a)$. However, deviations form the power law occur at high packing fractions. Because there are few points in the region where these deviations appear, it is not possible to conclude whether the curve is better described by a power law or an exponential function or maybe another form for high packing fractions.
Figure 2.11: Top panel: The dynamical correlation length $\xi(\tau_\alpha)$ and the dynamical susceptibility $\chi_4(\tau_\alpha)$ as functions of the packing fraction $\phi$, fitted to inverse power laws $\xi(\tau_\alpha) \propto (\phi_j - \phi)^{-\gamma_\xi}$ and $\chi_4(\tau_\alpha) \propto (\phi_j - \phi)^{-\gamma_\chi}$, respectively. Bottom panel: $\chi_4(\tau_\alpha)$ against $\xi(\tau_\alpha)$. The dashed line corresponds to the fit $\chi_4(\tau_\alpha) \approx \mathcal{A}(\xi(\tau_\alpha))^{d-p}$, with $d - p = 1.63$. 
2.2.3 Influence of the Parameter $a$ from the Correlation Function on $\xi$

All previous results were calculated using $a = 0.6 \ r_1$ in the Eq. 2.6. This section will be dedicated to analyze the dependence of $\chi_4(\tau_\alpha)$ and $\xi(\tau_\alpha)$ on $a$.

It has been found in experiments that the height of the peak of $\chi_4(t)$ increases with increasing values of $a$ up to a certain value, which will be called $a_{\text{max}}$. Then, for $a > a_{\text{max}}$ the peak is seen to decrease [41]. Commonly, the value of $a$ used to calculate the extent of the dynamical heterogeneity $\xi(t)$ is chosen to be $a = 0.6r_1$ [24, 29, 50]. However, the dependence of the correlation length $\xi(t)$ on the parameter $a$ has not been explored in detail.

The growing behavior of $\xi(\tau_\alpha)$ and $\chi_4(\tau_\alpha)$ as a function of $a$ is shown in Fig. 2.13 for the range $0.2r_1 \leq a \leq 4.0r_1$. It can be observed that, qualitatively, the behavior of the two quantities is very alike. Both grow with $a$ and go through three different regimes. First a
rapidly increase can be identified for \( a_1 \lesssim r_1 \), then, a crossover, and finally, a much slower growth for \( a_1 \gtrsim 1.2r_1 \).

Figure 2.13: *Top panel:* \( \xi(\tau_a) \) against \( a \) for \( \phi = 0.60, 0.65, 0.70, 0.72, 0.74, 0.76, 0.78, 0.79, 0.80 \) and 0.805 (from bottom to top). First, the growth of \( \xi(\tau_a) \) occurs very rapidly for small \( a \), then it goes through a crossover to finally go to a slower growth. *Bottom panel:* Growing behavior of \( \chi_4(\tau_a) \) as a function of \( a \) for the same packing fractions as in the top panel. \( \chi_4(\tau_a) \) grows very similarly to \( \xi(\tau_a) \) as a function of \( a \).
Furthermore, the results obtained in Fig. 2.11 for the value \( a = 0.6r_1 \) will be compared to two other choices of \( a \), namely \( a = 1.4r_1 \) and \( a = 3.0r_1 \). These results can be observed in Fig. 2.14 for \( a = 0.6r_1, 1.4r_1 \) and \( 3.0r_1 \). In the top panel of this figure a similar trend in the growing behavior of \( \xi(\tau_a) \) with \( \phi \) can be seen for the different values of \( a \). First, in the curve, the points of \( \xi(\tau_a) \) are very close to each other for low packing fractions, but then, the points start to differ considerably from each other for high packing fractions. This suggests that for the case of high packing fractions, when the heterogeneities in the dynamics become more pronounced, the selection of \( a \) has a big impact on the result of \( \xi \) than for low packing fractions, when the dynamics is governed by collisions between pairs of particles.

With this in mind, it should be interesting to determine how the relationship 
\[
\chi_4(\tau_a) \approx A \xi(\tau_a)^{d-p}
\]
changes with different values of \( a \). It turns out that despite the large difference in the values of \( \xi \) for high packing fractions with the choice of \( a \), the relationship \( \chi_4(\tau_a) \approx A \xi(\tau_a)^{d-p} \) only changes by the multiplicative constant \( A \), with the exponent \( d - p \) similar for all values of \( a \) mentioned before. These results are shown in the bottom panel of Fig. 2.14 for the three different values of \( a \). These results indicate that independently of the choice of \( a \), \( \chi_4(\tau_a) \) grows with \( \xi_4(\tau_a) \) in the same way.

For the packing fractions studied in this section the value of \( a_{\text{max}} \) was not reached. In order to reach this value, the simulations would have to be extended to much longer times. This is a difficult task considering the large number of collisions involved. It would be interesting to investigate Fig. 2.14 and Fig. 2.13 at \( a \approx a_{\text{max}} \) and \( a > a_{\text{max}} \). This suggestion is left for future investigation. My speculation is that \( \chi_4(\tau_a) \approx A \xi(\tau_a)^{d-p} \), even for \( a \geq a_{\text{max}} \), gives similar values of \( d - p \) to the ones I have already determined. This would mean that \( \xi(\tau_a) \) as a function of \( a \) peaks at \( a_{\text{max}} \) and then decreases for \( a > a_{\text{max}} \) (see Fig. 2.13 for reference).
Figure 2.14: **Top panel:** The dynamical correlation length $\xi(\tau_a)$ for different values of the parameter $a$ of the overlap function. The deviation of the curves becomes stronger for higher packing fractions. **Bottom panel:** $\chi_4(\tau_a)$ against $\xi(\tau_a)$ for three different choices of the parameter $a$. The different lines correspond to the fit $\chi_4(\tau_a) \approx A\xi(\tau_a)^{d-p}$ for each $a$. I find similar fitted values of $d - p$ for the three different values of $a$.

### 2.2.4 The Correlation Length $\xi$ as a Function of Time

In this section I turn to analyze the dependence of $\xi$ on time for a fixed packing fraction. It has been found in glasses that the typical lifetime of dynamical heterogeneity
is longer than the alpha relaxation time $\tau_\alpha$ [29, 53] and that $\xi(t)$ grows monotonically as a function of time for times less than $\tau_\alpha$. The behavior of the heterogeneous regions at longer times is rather controversial. While some research groups have found that the length of the regions stays constant [29] others have observed that the correlation length decays [24].

An analysis of the correlation length $\xi(t)$ as a function of time $t$ for this system can be seen in Fig. 2.15. This figure displays this dependence for the packing fractions $\phi = 0.74$, 0.76, 0.78 and 0.79. These correlation lengths are determined in the same way as in Sec. 2.2.2, with $a = 0.6r_1$ and $\varepsilon = 0.9$. $S_4(q, \tau_\alpha)$ was fitted to an OZ form for $q$ up to the value $q_m = 0.2$. For all packing fractions, the correlation length increases monotonically with time for times $t < 2\tau_\alpha$. For times $t > 2\tau_\alpha$, it was found that the data could not be well described by any of the fitting function presented in Sec. 2.2.2 (see Eqs. 2.10 and 2.11).

![Figure 2.15](image.png)

Figure 2.15: The correlation length $\xi$ against $t/\tau_\alpha$ for packing fractions $\phi = 0.79$, 0.78, 0.76 and 0.74. The correlation length $\xi(t)$ increases monotonically with time for $t < 2\tau_\alpha$. 
Because all fits were far off from the data points, the analysis at longer times than \( t > 2\tau_\alpha \) is excluded in this work and therefore, excluded in Fig. 2.15.

### 2.2.5 Correlation Length \( \xi \) for Different Degree of Inelasticity \( \varepsilon \)

In experiments in granular systems, the dissipation of energy through collisions is unavoidable as well as difficult to account for. Therefore, it is of high interest to know how the strength of the dissipation affects the nature of the dynamical heterogeneity. In these simulations, the degree of dissipation is controlled by \( \varepsilon \) (see Eq. (2.1)). In this section, the effects of having different values of \( \varepsilon \) are explored by analyzing \( \chi_4(\tau_\alpha) \) and \( \xi(\tau_\alpha) \) at different values of \( \varepsilon \). All results shown in previous sections were calculated with \( \varepsilon = 0.90 \).

The top panel of Fig. 2.16 shows, for the particular case of \( \phi = 0.76 \), the dynamical susceptibility \( \chi_4(t) \) for \( \varepsilon = 1.00, 0.90, 0.80 \) and 0.70. It can be observed that the height of the peak of \( \chi_4(t) \) increases with increasing \( \varepsilon \) and shifts to longer times. The lower panel of Fig. 2.16 show that \( \xi(\tau_\alpha) \) also grows as a function of the coefficient of restitution \( \varepsilon \). The growing behavior of both \( \chi_4 \) and \( \xi \) is compatible with an \( \varepsilon \)-dependent critical density \( \phi_j(\varepsilon) \) found in [54]. Figure 2.17 displays the results of \( \chi_4(\tau_\alpha) \) as a function of \( \xi(\tau_\alpha) \) obtained with the values of the coefficients of restitution \( \varepsilon = 1.0, 0.90, 0.8 \) and 0.70. The different points for a particular \( \varepsilon \) correspond to the packing fractions \( \phi = 0.72, 0.74, 0.76, 0.78 \) and 0.79. Surprisingly, the points for all coefficient of restitution and all packing fraction can be fitted to a common curve \( \chi_4(\tau_\alpha) \approx A\xi(\tau_\alpha)^{d-p} \), with \( d-p \approx 1.59 \). Therefore, it can be conclude that the relationship between \( \chi_4(\tau_\alpha) \) and \( \xi(\tau_\alpha) \) is independent of the coefficient of normal restitution \( \varepsilon \).

### 2.3 Discussion

In summary, in this chapter dynamical heterogeneity was studied in a two–dimensional driven granular fluid. In particular, the correlation length \( \xi(t) \), obtained from the four–point correlation function \( S_4(q,t) \), and the dynamical susceptibility \( \chi_4(t) \)
Figure 2.16: Top panel: $\chi_4(t)$ for packing fraction $\phi = 0.76$ calculated for systems with the coefficients of restitution $\varepsilon = 1.00, 0.90, 0.80$ and $0.70$. By increasing $\varepsilon$ the peak of $\chi_4(t)$ increases and shifts to longer times. Bottom panel: Correlation length $\xi(\tau_0)$ as a function of $\varepsilon$, for $\phi = 0.72, 0.74, 0.76, 0.78$, and $0.79$.

were studied as functions of packing fraction and time. The results were also compared for different choices of the parameter $a$ of the overlap function. Also, a more extensive analysis was performed by comparing the results of $\xi(\tau_0)$ and $\chi_4(\tau_0)$ for different values of the coefficient of restitution $\varepsilon$ which determines the energy dissipation in the system.
Figure 2.17: $\chi_4(\tau_\alpha)$ against $\xi(\tau_\alpha)$ fitted to $\chi_4(\tau_\alpha) \approx A\xi(\tau_\alpha)^{d-p}$ (dotted–dashed points) for the values $\epsilon = 1.00, 0.90, 0.80$ and $0.70$. The different points for a $\epsilon$ value correspond to the packing fractions $\phi = 0.72, 0.74, 0.76, 0.78,$ and $0.79$. The values $d-p = 1.59$ was obtained by fitting all points together.

The dynamical susceptibility $\chi_4(\tau_\alpha)$ and correlation length $\xi(\tau_\alpha)$ were observed to increase with increasing packing fraction $\phi$ toward the jamming transition $\phi_J$.

Additionally, the values of $\xi(\tau_\alpha)$ for high packing fraction found in this granular fluid appear to be much larger than in any other system studied before [15, 24, 29, 41, 49]. For this system the dependence of the relaxation time $\tau_\alpha$ on the packing fraction $\phi$ was better fitted to an exponential form than to a power law in the whole $\phi$ range, in agreement with results found in [47, 48]. Qualitatively, the growing behavior of $\chi_4(\tau_\alpha)$ and $\xi(\tau_\alpha)$ with $\phi$ agrees with experimental results obtained by using a driving method comparable to the one used here [25]. It was also found that $S_4(q, \tau_\alpha)$ is well described by an Ornstein-Zernicke fit only for small values of $q < 3$. However, the four point dynamic structure factor is well described by a scaling form $S_4(q, \tau_\alpha)/\chi_4(\tau_\alpha) = s[q\xi(\tau_\alpha)]$ over the
whole range of $\xi q$, as long as $|q| < 0.3$. Also, the results for $\xi(\tau_\alpha)$, obtained from fitting the four point correlation function, $S_4(q, \tau_\alpha)$, only vary slightly with different fitting functions and with different fitting ranges $[0, q_m]$. It is also important to remark that the values of $\chi_4(t)$, obtained by calculating the fluctuations of the two point correlation function $Q(t)$, do correspond to the limit $\lim_{q\to 0} S_4(q, t)$. This is true in this case because the particle concentration and the packing fraction are allowed to fluctuate. Therefore, $\chi_4(t)$ accounts for all fluctuations. Furthermore, both $\xi(\tau_\alpha)$ and $\chi_4(\tau_\alpha)$ were well fitted by mode-coupling-like functions $\xi(\tau_\alpha) \propto (\phi_J - \phi)^{-\gamma_\xi}$ and $\chi_4(\tau_\alpha) \propto (\phi_J - \phi)^{-\gamma_\chi}$, with exponents $\gamma_\xi = 1.32$ and $\gamma_\chi = 2.04$, respectively. The relationship between the dynamical susceptibility and the correlation length is given by $\chi_4(\tau_\alpha) \approx \mathcal{A}(\xi(\tau_\alpha))^{d-p}$, where $d-p \approx 1.63$ can be interpreted as the fractal dimension of the correlated regions.

The relation between the growing time scales and length scales can be described with a power law for low packing fraction $\phi < 0.76$. However, for higher packing fractions the form of the relation between $\tau_\alpha$ and $\xi(\tau_\alpha)$ could not be determined with precision due to limitations in the data.

It was observed that the dynamical susceptibility $\chi_4(\tau_\alpha)$ and the correlation length $\xi(\tau_\alpha)$ are increasing function of the cut–off length $a$ in the overlap function. The growing behavior of both quantities goes through three different regimes in the range $0.2r_1 < a < 4.0r_1$. The relationship $\chi_4(\tau_\alpha) \approx \mathcal{A}(\xi(\tau_\alpha))^{d-p}$ was found to hold for all values of $a$, and the exponent $d-p \approx 1.63$ was found to take approximately the same value for different values of $a$. The only dependence on $a$ appears in the multiplicative constant $\mathcal{A}(a)$.

$\xi$ was found to grow monotonically with $t$ for $t < 2\tau_\alpha$. Unfortunately, the results of $t > 2\tau_\alpha$ are inconclusive. It was found that none of the fitting functions presented here described the data for long times.
Finally, as the coefficient of restitution $\varepsilon$ is increased, the height of the peak of $\chi_4(t)$ increases and the position of the peak shifts to longer times. Moreover, the relationship $\chi_4(\tau_\alpha) \approx A(\xi(\tau_\alpha))^{d-\mu}$ was found to be insensitive to the degree of inelasticity of the particle collisions.

Altogether, the results obtained in this chapter suggest that the geometry of the clusters is insensitive to the choice of cutoff parameter $a$ and to the degree of inelasticity of the collisions between the particles. Also, in general, the quantities $\tau_\alpha$, $\chi_4$ and $\xi$ increase very rapidly as the jamming transition is approached, and they show qualitatively similar behavior compared to simulations and experiments in other glassy systems. However, the highest values of $\xi$ found here appear to be significantly larger than the ones found in any previous experiment or simulation in glassy systems [15, 24, 29, 41, 49].
3  **PROJECT 2: TESTING THE PRESENCE OF TIME**

**REPARAMETRIZATION INVARIANCE IN STRUCTURAL GLASSES**

In this chapter I use triangular relations (Sec. 1.5.4.1) to test the theoretical framework presented in Sec. 1.5.4 that proposes an explanation for the origin of dynamical heterogeneity in structural glasses. The tests are applied to data from the numerical simulations described in Sec. 3.1. Most of the results presented in this chapter were published in [55, 56]

In this project, the analysis requires one to coarse grain the system in order to quantify the local fluctuations. The coarse graining has to fulfill certain conditions that are briefly explained in Sec. 3.1.4. The triangular relations, described in Sec. 1.5.4.1, are studied with two different approaches. In Sec. 3.2.1, I introduce the results from an analysis inspired by the one proposed in Ref. [33], which is also described in Sec. 1.5.4.1. In Sec. 3.2.2, I present a modified version of the method presented in Sec. 3.2.1. In this second method, the discrimination between the time reparametrization fluctuations and other fluctuations can be done more simply. Furthermore, a more quantitative analysis that directly compares the time reparametrization fluctuations with those related to dynamical heterogeneities is presented in Sec. 3.3. All mathematical quantities required for the aforementioned analysis are derived in the same section. At the end of this chapter, I summarize all the results and discuss them in terms of the time reparametrization invariance hypothesis.
3.1 Models and Simulation Details

3.1.1 Lennard-Jones (LJ) Potential

The Lennard–Jones potential is a simple model utilized to describe interactions between molecules. The potential has the functional form

\[ V_{LJ}(r) = 4\epsilon \left[ \left( \frac{\sigma}{r} \right)^{12} - \left( \frac{\sigma}{r} \right)^6 \right], \]

where \( \epsilon \) is the depth of the potential well and \( \sigma \) gives the order of magnitude of the distance at which the potential changes from being attractive to repulsive. Because of the steep repulsive wall that arises for \( 0 < r < \sigma \), \( \sigma \) is sometimes regarded as a rough measure of the diameter of the molecules. This potential is shown in Fig. 3.1. The natural temperature unit for a potential of this form is \( \tilde{T} = \epsilon/k_B \), where \( k_B \) is Boltzmann’s constant.

The computational load of the simulations involving this potential is usually reduced by the choice of a cutoff distance of \( r^{(\text{cutoff})} = 2.5\sigma \). Each particle only interacts with

![Figure 3.1: Lennard-Jones potential described in Eq. (3.1).](image-url)
particles at distances smaller than this cutoff length. Moreover, in order to avoid crystallization at low temperatures in simulations of glassy systems it is commonly assumed that there are two types of particles, here labeled as A and B. With these conditions the potential is expressed as follows:

\[
V_{LJ_{\alpha\beta}}(r) = 4\epsilon_{\alpha\beta} \left[ \left( \frac{\sigma_{\alpha\beta}}{r} \right)^{12} - \left( \frac{\sigma_{\alpha\beta}}{r} \right)^{6} \right] \theta(r^{(\text{cutoff})}_{\alpha\beta} - r),
\]  

(3.2)

where \(\alpha\) and \(\beta \in \{A, B\}\) and \(\theta(x)\) is the Heaviside theta function [57, 58].

### 3.1.2 Weeks-Chandler-Andersen (WCA) Potentials

In some cases, it is useful to separate the attractive and repulsive parts of the Lennard–Jones potential. For this purpose, the following functional form was proposed by Weeks, Chandler and Andersen [59, 60]:

\[
V_{WCA} = \begin{cases} 
V_{LJ} + \epsilon & ; \quad r < r_{\text{min}} \\
0 & ; \quad r \geq r_{\text{min}} 
\end{cases}
\]

(3.3)

where \(r_{\text{min}} = 2^{1/6}\sigma\) is the position of the minimum of \(V_{LJ}\). This potential, shown in Fig. 3.2, is the repulsive part of \(V_{LJ}\). Similarly to Eq. 3.4, in the case of bidisperse systems, the potential can be written as follows

\[
V_{WCA_{\alpha\beta}}(r) = 4\epsilon_{\alpha\beta} \left[ \left( \frac{\sigma_{\alpha\beta}}{r} \right)^{12} - \left( \frac{\sigma_{\alpha\beta}}{r} \right)^{6} + 1 \right] \theta(r^{(\text{cutoff})}_{\alpha\beta} - r),
\]

(3.4)

where the cut-off distance is set at \(r^{(\text{cutoff})}_{\alpha\beta} = 2^{1/6}\sigma_{\alpha\beta} \approx 1.1225\sigma_{\alpha\beta}\).

### 3.1.3 Simulation Details

All the raw data analyzed in this work were obtained in simulations by Dr. Azita Parsaeian [26, 28, 37, 61]. The classical molecular dynamics simulations were performed in systems of \(N\) particles that were initially equilibrated at high temperature \(T_i \gg T_c\) (where \(T_c\) is the critical temperature predicted by mode coupling theory [3]), then
Figure 3.2: The WCA Potential described in Eq. (3.3). This potential consists of the repulsive part of the LJ potential.

instantaneously quenched to a final temperature $T$ and allowed to evolve for times several orders of magnitude longer than their typical vibrational times [26, 28, 37, 61]. In total, eight data sets were generated by simulating four atomistic glass-forming models [61]. Two of the systems are 80:20 mixtures of A and B particles, interacting via either Lennard-Jones (LJ) potentials [57, 58] or via purely repulsive Weeks-Chandler-Andersen (WCA) potentials [59, 60]. The interactions in the particle systems have the same length parameters, $\sigma_{\alpha\beta}$ ($\alpha, \beta \in \{A, B\}$), and energy parameters, $\epsilon_{\alpha\beta}$ as in the standard Kob-Andersen mixture, namely [57, 58]: $\sigma_{AA} = 1.0$, $\sigma_{AB} = 0.8$, $\sigma_{BB} = 0.88$, $\epsilon_{AA} = 1.0$, $\epsilon_{AB} = 1.5$ and $\epsilon_{BB} = 0.5$. The other two systems are models of short (10-monomer) polymers, in which all particles interact with each other via either LJ potentials or via WCA potentials, with length parameter $\sigma_{AA} = 1.0$ and energy parameter $\epsilon_{AA} = 1.0$. Additionally, in the polymer models, nearest-neighbor monomers along a chain are connected by a finite extendable nonlinear elastic (FENE) anharmonic spring
potential [62]. As discussed before, for both particle and polymer systems the LJ potential is truncated at the cut-off distance \( r_{\text{cutoff,}\alpha\beta} = 2.5\sigma_{\alpha\beta} \) and the WCA potential is truncated at \( r_{\text{cutoff,}\alpha\beta} = 2^{1/6}\sigma_{\alpha\beta} \). The unit of length is chosen as \( \sigma_{AA} \), the unit of energy as \( \epsilon_{AA} \) and the unit of time as \( (\sigma_{AA}^2 M/48\epsilon_{AA})^{1/2} \). For the particle systems, the simulations were performed in an NVT ensemble, with the temperature being fixed by the rescaling method [63]. For the polymer systems, the simulations were performed in an NPT ensemble, with both the pressure and the temperature being controlled by the Nose-Hoover method [64, 65]. The details of the simulations are summarized in Table 3.1.

Table 3.1: Details of the numerical simulations that generated the data analyzed in this work. The simulations were performed by Dr. Azita Parsaeian. Each system consists of \( N \) particles, with \( m \) particles per molecule, interacting via either Lennard–Jones (LJ) potentials or via purely repulsive Weeks–Chandler–Andersen (WCA) potentials, at final temperature \( T \). Each temperature is also described by its ratio with respect to the empirically determined mode coupling critical temperature \( T_c \) [3] for the same system. The last column lists the number of independent runs. Data sets F and H correspond to systems in equilibrium, all others to systems in the aging regime.

<table>
<thead>
<tr>
<th>Label</th>
<th>( N )</th>
<th>( m )</th>
<th>Potential</th>
<th>( T )</th>
<th>( T_c )</th>
<th>( T/T_c )</th>
<th>Runs</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>8000</td>
<td>10</td>
<td>LJ</td>
<td>0.6</td>
<td>0.833</td>
<td>( \approx 0.7 )</td>
<td>100</td>
</tr>
<tr>
<td>B</td>
<td>8000</td>
<td>10</td>
<td>WCA</td>
<td>0.4</td>
<td>0.503</td>
<td>( \approx 0.8 )</td>
<td>800</td>
</tr>
<tr>
<td>C</td>
<td>8000</td>
<td>1</td>
<td>LJ</td>
<td>0.4</td>
<td>0.435</td>
<td>( \approx 0.9 )</td>
<td>250</td>
</tr>
<tr>
<td>D</td>
<td>1000</td>
<td>1</td>
<td>WCA</td>
<td>0.236</td>
<td>0.263</td>
<td>( \approx 0.9 )</td>
<td>5000</td>
</tr>
<tr>
<td>E</td>
<td>1000</td>
<td>1</td>
<td>WCA</td>
<td>0.29</td>
<td>0.263</td>
<td>( \approx 1.1 )</td>
<td>9000</td>
</tr>
<tr>
<td>F</td>
<td>1000</td>
<td>1</td>
<td>WCA - Eq</td>
<td>0.29</td>
<td>0.263</td>
<td>( \approx 1.1 )</td>
<td>9000</td>
</tr>
<tr>
<td>G</td>
<td>1000</td>
<td>1</td>
<td>WCA</td>
<td>0.4</td>
<td>0.263</td>
<td>( \approx 1.5 )</td>
<td>4999</td>
</tr>
<tr>
<td>H</td>
<td>1000</td>
<td>1</td>
<td>WCA - Eq</td>
<td>0.4</td>
<td>0.263</td>
<td>( \approx 1.5 )</td>
<td>4999</td>
</tr>
</tbody>
</table>
The mode coupling critical temperature $T_c$ was determined by fitting $\tau_\alpha = (T - T_c)^{-\gamma}$, where $\tau_\alpha$ is the equilibrium $\alpha$-relaxation time defined by the conditions $C(t_w + \tau_\alpha(t_w), t_w) = 1/e$ (see Eq. 1.2) and $\tau_\alpha = \lim_{t_w \to \infty} \tau_\alpha(t_w)$, as shown in Fig. 3.3 for the WCA and LJ polymers (see also Ref. [26]).

![Figure 3.3](image)

Figure 3.3: Alpha relaxation time $\tau_\alpha$ as a function of temperature for the WCA and LJ polymers. The dashed line represents the fit $\tau_\alpha \propto (T - T_c)^{-\gamma}$. The fits estimated $T_c = 0.50$ and $\gamma = 1.22$ for the WCA polymers and $T_c = 0.83$ and $\gamma = 1.88$ for the LJ polymers.

### 3.1.4 Averaging and Coarse Graining

In this chapter, the meaning of the notations $\langle \cdots \rangle$ and $\cdots$ differ from Chapter 2. Here I use $\langle \cdots \rangle$ to denote an average over thermal fluctuations, which in our case is approximated by an average over independent molecular dynamics (MD) runs, and $\cdots$ to denote a spatial average.

To select an appropriate coarse-graining for the analysis, the behavior of the contours of the constant probability density of the local correlations were observed for different
coarse-graining sizes. In order to coarse-grain, the system was divided into regions. These regions have the same size, but may slightly differ in the number of particles they contain. I found that the contours show a clearer anisotropy when the average number of particles contained in each box is approximately 125. Thus, unless otherwise indicated, every analysis of the local fluctuations performed in this work is calculated with regions that contain on average 125 particles. The reason why the coarse-grained regions must accommodate at least this amount of particles can be explained in terms of the longitudinal and transverse fluctuations [34]. Here, the Goldstone modes associated with the broken continuous symmetry correspond to transverse fluctuations. All other fluctuations are longitudinal fluctuations. It is known that in general transverse fluctuations are more correlated over long distances than longitudinal fluctuations [34]. Thus, for larger coarse-graining boxes longitudinal fluctuations in different points of the box will be weakly correlated and will tend to cancel each other. The larger the box the more the transverse fluctuations are expected to dominate.

3.2 Results

3.2.1 First Analysis of Time Reparametrization Fluctuations

3.2.1.1 Global Quantities

In this section, the hypothesis of time reparametrization invariance, given by Eq. (1.10), is tested by using the triangular relations presented in Sec. 1.5.4.1. Because the data scales as shown in Eq. (1.12) [5], the function \( f \) can be determined by fitting the resulting \( C(t, t_w) \) versus \( h(t)/h(t_w) \) curve. The different forms of \( h(t) \) found for the systems studied here are: for the aging polymers \( h(t) = \exp[\ln^\alpha(t/t_0)] \), for the aging particles \( h(t) = \exp[(t/t_0)^\alpha] \), and for the equilibrated systems \( h(t) = \exp(t/t_0) \). These different forms of \( h(t) \) were found to be dependent on the details of the relaxation process [5]. This means
that the dependence of the \( \alpha \) relaxation time \( \tau_\alpha \) on the waiting time \( t_w \) differs considerably for the different systems (See Fig. 3.4).

The result of plotting \( C(t, t_w) \) against \( h(t)/h(t_w) \) is shown in Fig. 3.5 for all systems. The function \( f \) can be fitted with a form such that \( C(t, t_w) \) reduces to a stretched exponential in the equilibrium case:

\[
f(x) = q_{EA} \exp\{-[\ln(x)/\theta_0]^\beta\}. \tag{3.5}
\]

In Fig. 3.5, the function \( f(x) \) shown as a solid line corresponds to the fitting parameters \( q_{EA} = 0.76, \beta = 0.82 \) and \( \theta_0 = 1 \). However, the function \( f(x) \) was fitted individually for each data set. All fitting parameters and the functional forms of \( h(t) \) are summarized in Table 3.2. The fitting parameters \( q_{EA}, \beta \) and \( \theta_0 \) vary little from one data set to another.

Figure 3.4: Relaxation time \( \tau_\alpha(t_1) \) as a function of the waiting time \( t_1 \) for all the models and temperatures considered. \( \tau_\alpha(t_1) \) is defined by the condition \( C(t_1 + \tau_\alpha(t_1), t_1) = 1/e \).
Figure 3.5: Scaling form of $C(t, t_w)$ vs $h(t)/h(t_w)$. All data sets approximately collapse with each other to the function $f(x) = q_{EA} \exp\{-(\ln(x)/\theta_0)^\beta\}$, with parameters $q_{EA} = 0.76$, $\beta = 0.82$ and $\theta_0 = 1$ (solid line).

This is expected because the curves $C(t, t_w)$ against $h(t)/h(t_w)$ for different data sets (shown in Fig. 3.5) do not differ significantly from each other.

Now that $f(x)$ is known, the global curve $1 = XY$ discussed in Sec. 1.5.4.1 can in principle be determined. These results are shown in Fig. 3.6 for all systems and all possible times $t_1 < t_2 < t_3$. The points for all data sets can be seen to be in good agreement with the relation $1 = XY$. Any departure from this relation is due to the fact that the fitting functions are not exactly identical to the $C(t, t_w)$ data. Even when the collapse to a single curve is not perfect, it is good enough to be used for a test of the time reparametrization hypothesis.
Table 3.2: Fitted functions $h(t)$ and $f(x)$ together with the fitting parameters found for all data sets.

<table>
<thead>
<tr>
<th>Label</th>
<th>$h(t)$</th>
<th>$f(x)$</th>
<th>$\alpha$</th>
<th>$t_0$</th>
<th>$q_{EA}$</th>
<th>$\beta$</th>
<th>$\theta_0$</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>$\exp(\ln^\alpha(t/t_0))$</td>
<td>$q_{EA} \exp[-(\ln(x)/\theta_0)\beta]$</td>
<td>0.53</td>
<td>2.88</td>
<td>0.78</td>
<td>0.83</td>
<td>0.93</td>
</tr>
<tr>
<td>B</td>
<td>$\exp(\ln^\alpha(t/t_0))$</td>
<td>$q_{EA} \exp[-(\ln(x)/\theta_0)\beta]$</td>
<td>0.88</td>
<td>2.88</td>
<td>0.74</td>
<td>0.81</td>
<td>1.46</td>
</tr>
<tr>
<td>C</td>
<td>$\exp((t/t_0)\alpha)$</td>
<td>$q_{EA} \exp[- \ln(x)^\beta]$</td>
<td>0.25</td>
<td>17.09</td>
<td>0.72</td>
<td>0.83</td>
<td>–</td>
</tr>
<tr>
<td>D</td>
<td>$\exp((t/t_0)\alpha)$</td>
<td>$q_{EA} \exp[- \ln(x)^\beta]$</td>
<td>0.16</td>
<td>0.802</td>
<td>0.76</td>
<td>0.81</td>
<td>–</td>
</tr>
<tr>
<td>E</td>
<td>$\exp((t/t_0)\alpha)$</td>
<td>$q_{EA} \exp[- \ln(x)^\beta]$</td>
<td>0.27</td>
<td>1.44</td>
<td>0.73</td>
<td>0.82</td>
<td>–</td>
</tr>
<tr>
<td>F</td>
<td>$\exp(t/t_0)$</td>
<td>$q_{EA} \exp[- \ln(x)^\beta]$</td>
<td>–</td>
<td>744.78</td>
<td>0.73</td>
<td>0.73</td>
<td>–</td>
</tr>
<tr>
<td>G</td>
<td>$\exp((t/t_0)\alpha)$</td>
<td>$q_{EA} \exp[- \ln(x)^\beta]$</td>
<td>0.3</td>
<td>0.14</td>
<td>0.79</td>
<td>0.86</td>
<td>–</td>
</tr>
<tr>
<td>H</td>
<td>$\exp(t/t_0)$</td>
<td>$q_{EA} \exp[- \ln(x)^\beta]$</td>
<td>–</td>
<td>24.53</td>
<td>0.69</td>
<td>0.82</td>
<td>–</td>
</tr>
</tbody>
</table>

Figure 3.6: Global $(\Psi_{21}/\sqrt{\Psi_{31}}, \Psi_{32}/\sqrt{\Psi_{31}})$ pairs for all data sets and all possible times $t_1 < t_2 < t_3$. The expected relation $1 = XY$ is shown with a dashed line.
3.2.1.2 Local Quantities

As discussed in Sec. 1.5.4.1, the \((X_r, Y_r)\) points calculated from local quantities are expected to be mostly located along the global curve shown in Fig. 3.6. In order to test this, one can calculate 2d projections of the joint probability density of the coarse-grained correlations. The results of the 2d projections,

\[ \rho(X_r, Y_r) = \rho(\Psi_{21,r}/(\Psi_{31,r})^{1/2}, \Psi_{23,r}/(\Psi_{31,r})^{1/2}) \],

are presented in Fig. 3.7 for all systems. The contours were computed using coarse graining regions containing 125 particles on average. In this figure, only the contours that enclose 25%, 50% and 75% of the total probability are shown. For each data set, I present the contours spread around the global points \((X(t_1, t_2, t_3), Y(t_1, t_2, t_3)) \approx (0.8, 1.25), (1.00, 1.00), \) and \((1.25, 0.80)\), with \(t_1 < t_2 < t_3\). Since time reparametrization symmetry is a long time asymptotic effect, the times \(t_1\) are chosen as late as possible. The results corresponding to data sets A-D, with \(T/T_c \approx 0.7 - 0.9\), present contours that are mostly concentrated along the global curve \(1 = XY\), as the time reparametrization theory predicts. Similar results were also found for spin glasses in Ref. [33]. The anisotropy is more noticeable for the 25% contour, which encloses the most likely fluctuations, than for the 50% and 75% contours, which additionally include rarer events. For data sets E and F, corresponding to \(T/T_c = 1.1\), the contours are still anisotropic and oriented along the direction of the global curve, but less so than in A-D. Lastly, for data sets G and H, corresponding to \(T/T_c = 1.5\) the fluctuations away from the global curve are the strongest. These results suggest that for large temperature, the effect of local time variable fluctuations become weaker.
Figure 3.7: 2D contours of constant joint probability density \( \rho(X_r, Y_r) = \rho(\Psi_{21,r}/(\Psi_{31,r})^{1/2}, \Psi_{23,r}/(\Psi_{31,r})^{1/2}) \), computed using coarse graining boxes containing 125 particles on average. Each panel from A to H contains results from the corresponding data set, for the global points \((X, Y) \approx (0.80, 1.25), (1.00, 1.00) \) and \((1.25, 0.80)\), with the times chosen as late as possible within each data set. Each set of three concentric contours is chosen so that they enclose 25%, 50% and 75% of the total probability.
It is surprising that even when the aging relaxation dynamics is different for systems A, B, C and D, the probability contours for local fluctuations are almost identical across systems as long as the times are chosen so that the global point \((X(t_1, t_2, t_3), Y(t_1, t_2, t_3))\) is the same for all four of them. However, the shapes of the contours at different points \((X(t_1, t_2, t_3), Y(t_1, t_2, t_3))\) are different. This effect may be an artifact of the curvature of the global curve. To compare the contours at different points of the global curve a better possibility would be if the global curve is a straight line rather then the curve \(1 = XY\). This will be the focus of the next section.

3.2.2 Second Analysis of Time Reparametrization Fluctuations

3.2.2.1 Global Quantities

A different global curve than the one obtained in the previous section could be generated such that it has the time reparametrization fluctuations, “transverse fluctuations”, along the x axis and the other fluctuations, “longitudinal fluctuations”, along the y axis, i.e., the new global curve would be a straight line at \(Y=0\). This would allow a better comparison between these two different kinds of fluctuations. Also, in this way, the shape of the contours of constant probability density the local fluctuations would not be affected by the curvature of the global curve.

To determine the new axes \(X\) and \(Y\) that give a straight line at \(Y = 0\), one could start from the previous relation \(1 = XY\) (see Sec. 1.5.4.1 and 3.2.1). By taking the natural logarithm of \(1 = XY\) one can directly obtain a straight line: \(\hat{X} + \hat{Y} = 0\), with \(\hat{X} \equiv \ln X = \ln \Psi_{32} - \frac{1}{2} \ln \Psi_{31}\) and \(\hat{Y} \equiv \ln Y = \ln \Psi_{21} - \frac{1}{2} \ln \Psi_{31}\). However, this line is not yet positioned at \(Y = 0\). To simplify the notation, I make the following change \(\varphi(t) = \ln(h(t))\). This, of course, introduces changes in other previously defined quantities, such as

\[
C(t, t_w) = g[\varphi(t) - \varphi(t_w)], \tag{3.6}
\]
and
\[ \Phi_{ab} = g^{-1}(C_{ab}) = \ln \Psi_{ab}, \] (3.7)

with \( a, b \in \{1, 2, 3\} \). With these changes, I proceed to the calculation of the new global curve. In order to move the straight line to the horizontal axis, the following transformation can be introduced \( \hat{U} = (\hat{X} - \hat{Y})/\sqrt{2} \) and \( \hat{V} = (\hat{X} + \hat{Y})/\sqrt{2} \). This corresponds to
\[ \hat{U} = \frac{\Phi_{32} - \Phi_{21}}{\sqrt{2}} \] (3.8)

and
\[ \hat{V} = \frac{\Phi_{32} + \Phi_{21} - \Phi_{31}}{\sqrt{2}}. \] (3.9)

Moreover, one can define an orthogonal matrix containing \( \hat{U} \) and \( \hat{V} \) in the following way:
\[ \sigma = \frac{1}{\sqrt{3}} (\Phi_{21} + \Phi_{32} - \Phi_{31}) = \sqrt{\frac{2}{3}} \hat{V}, \] (3.10)

\[ \pi_1 = \frac{1}{\sqrt{2}} (\Phi_{21} - \Phi_{32}) = -\hat{U}, \] (3.11)

and
\[ \pi_2 = \frac{1}{\sqrt{6}} (\Phi_{21} + \Phi_{32} + 2\Phi_{31}), \] (3.12)

with \( t_1 < t_2 < t_3 \). If the global two-time function verifies the relation in Eq. (3.6) then
\[ \sigma = \frac{1}{3} \left[ g^{-1}[C(t_2, t_1)] + g^{-1}[C(t_3, t_2)] - g^{-1}[C(t_3, t_1)] \right] = 0. \] (3.13)

By identifying the functions \( g(x) \) and \( \phi(t) \) from the forms of \( f(x) \) and \( h(t) \), obtained in Sec. 3.2.1, then the global curves can be computed directly. For the aging polymer
systems $\varphi(t) = \ln^\alpha(t/t_0)$, for the aging particle systems $\varphi(t) = (t/t_0)^\alpha$, and for the equilibrated systems $\varphi(t) = t/t_0$. Also,

$$\Phi_{ab} = g^{-1}[C(t_a, t_b)] = \theta_0[-\ln[q_{EA}^{-1}C(t_a, t_b)]]^{1/\beta},$$

(3.14)

where the values of the parameters $q_{EA}, \beta$ and $\theta_0$ are already known (see Table 3.2). In Fig. 3.8, the global values of $\sigma$ are plotted against the global values of $\sqrt{\pi_1^2 + \pi_2^2}$ for all times, $t_1 < t_2 < t_3$, and all systems. The collapse shown in this figure is good enough for the hypothesis to be tested by using these data and fits.

![Figure 3.8: Global ($\sigma, \sqrt{\pi_1^2 + \pi_2^2}$) pairs for all data sets and all possible times $t_1 < t_2 < t_3$.](image)

3.2.2.2 Local Quantities

To analyze the local fluctuations in the dynamics one can define a set of local quantities that are analogous to the global ones defined in Sec. 3.2.2.1.

If the local fluctuations were mostly due to Goldstone modes, they should satisfy

$$C_r(t, t_w) \approx g[\varphi_r(t) - \varphi_r(t_w)].$$

(3.15)
In principle, the functions $g(x)$ in Eq. (3.15) and Eq. (3.6) could be different. For example, it has been claimed [14] that stretched–exponential global relaxation could be the result of combining local exponential relaxations with different relaxation times, in which case we would have $g_{\text{global}} = A \exp (-|x|^\beta)$ in Eq. (3.6) and $g_{\text{local}} = A' \exp (-|x|)$ in Eq. (3.15) [40]. In this work, for simplicity, the condition $g_{\text{global}}(x) = g_{\text{local}}(x) = g(x)$ is imposed. Both this restriction and the restriction imposed in Eq. (3.6) could in principle make the results appear to be slightly less consistent with the hypothesis than they would be otherwise.

To perform the local analysis I start by defining

$$\Phi_{ab,\vec{r}} \equiv g^{-1}(C_{ab,\vec{r}}),$$

(3.16)

with $a, b \in \{1, 2, 3\}$, where $C_{ab,\vec{r}} \equiv C_r(t_a, t_b)$. The fluctuating quantity $\Phi_{ab,\vec{r}}$ is composed of a transverse contribution,

$$\Phi_{ab,\vec{r}}^T = \phi_r(t_a) - \phi_r(t_b),$$

(3.17)

and a longitudinal contribution, $\Phi_{ab,\vec{r}}^L$, i.e.

$$g^{-1}(C_{ab,\vec{r}}) = \Phi_{ab,\vec{r}} = \Phi_{ab,\vec{r}}^T + \Phi_{ab,\vec{r}}^L = \phi_r(t_a) - \phi_r(t_b) + \Phi_{ab,\vec{r}}^L.$$  

(3.18)

This means that in the absence of longitudinal fluctuations, Eq. (3.15) would be exact. In order to quantify both kinds of fluctuations, I perform a similar transformation to the one I did for the global variables:

$$\sigma_{\vec{r}} \equiv \frac{1}{\sqrt{3}} \left[ g^{-1}(C_{21,\vec{r}}) + g^{-1}(C_{32,\vec{r}}) - g^{-1}(C_{31,\vec{r}}) \right]$$

$$= \frac{1}{\sqrt{3}} \left( \Phi_{21,\vec{r}}^L + \Phi_{32,\vec{r}}^L - \Phi_{31,\vec{r}}^L \right),$$

(3.19)

$$\pi_{1,\vec{r}} \equiv \frac{1}{\sqrt{2}} \left[ g^{-1}(C_{21,\vec{r}}) - g^{-1}(C_{32,\vec{r}}) \right]$$

$$= \frac{1}{\sqrt{2}} \left[ (\Phi_{21,\vec{r}}^T + \Phi_{21,\vec{r}}^L) - (\Phi_{32,\vec{r}}^T + \Phi_{32,\vec{r}}^L) \right].$$

(3.20)
and

\[\pi_{2, r} \equiv \frac{1}{\sqrt{6}} \left[ g^{-1}(C_{21, r}) + g^{-1}(C_{32, r}) + 2g^{-1}(C_{31, r}) \right] \]

\[= \frac{1}{\sqrt{6}} \left[ (\Phi_{21, r} + \Phi_{21, r}^T) + (\Phi_{32, r} + \Phi_{32, r}^T) + 2(\Phi_{31, r} + \Phi_{31, r}^T) \right] , \quad (3.21)\]

with \( t_1 < t_2 < t_3 \). As shown above, \( \sigma_{r} \) contains only longitudinal fluctuations, but \( \pi_{1, r} \) and \( \pi_{2, r} \) contain both transverse and longitudinal components. If Eq. (3.15) was an exact identity, the local two-time function would verify the following relation

\[\sigma_{r} = \frac{1}{\sqrt{3}} \left[ g^{-1}(C_{21, r}) + g^{-1}(C_{32, r}) - g^{-1}(C_{31, r}) \right] = 0. \quad (3.22)\]

Therefore, in the case where no longitudinal fluctuations are present, the vector \((\sigma_{r}, \pi_{1, r}, \pi_{2, r})\) would be restricted to be fluctuating in the plane \(\sigma_{r} = 0\). It is expected that as the temperature becomes lower, the timescales become longer, and the system becomes more glassy, transverse fluctuations should become progressively more dominant, according to the hypothesis (Eq. (1.10)) and according to the observations in the previous section. Therefore, the probability distribution \(\rho(\sigma_{r}, \pi_{1, r}, \pi_{2, r})\) should become anisotropic, extending mostly along the \(\sigma = 0\) plane and not away from it. Figure 3.9 shows the results of plotting, for all systems, the 2D projection of the joint probability density of the coarse-grained local correlations, \(\rho(|\Delta\sigma_r|, |\Delta\pi_r|)\), with \(|\Delta\sigma_r| \equiv |\sigma_{r} - \sigma|\) and \(|\Delta\pi_r| \equiv \sqrt{(\pi_{1, r} - \pi_{1})^2 + (\pi_{2, r} - \pi_{2})^2}\). The values of the global quantities are subtracted from the local quantities to avoid trivial effects due to differences in the global values. By doing this it is possible to better compare the contours independently of the choice of \(C(t_2, t_1)\) and \(C(t_3, t_2)\). This kind of comparison was not possible in the analysis presented in Sec. 3.2.1.2 since the shape of the contours were affected by the curvature of the global curve. The value of \(C(t_3, t_1)\) is approximately the same for all systems, in this case \(C(t_3, t_1) \approx 0.23\). The three contours shown for each data set enclose respectively 25%,
50% and 75% of the total probability. As in the case of the results of Sec. 3.2.1, the anisotropy of the contours are qualitatively similar for all systems. For data sets with $T < T_c$ (A-D), the purely longitudinal fluctuations $\Delta \sigma_{\vec{r}}$ are clearly smaller than the $\Delta \vec{\pi}_{\vec{r}}$ fluctuations, which contain both transverse and longitudinal contributions. For moderately higher temperature, $T/T_c \approx 1.1$ (data sets E and F), the anisotropy $|\Delta \sigma_{\vec{r}}| < |\Delta \vec{\pi}_{\vec{r}}|$ is still present, but less pronounced. In the case of the highest temperature, $T/T_c \approx 1.5$ (data sets G and H), the anisotropy is either very slight, or absent. In the case of the systems that are equilibrated, F and H, it was found that the shapes of their contours are similar but slightly more anisotropic than the ones obtained for the same temperature in the aging regime, E and G, respectively.

The effect of temperature in the anisotropy of the contours discussed above can be observed in more detail in Fig. 3.10(a). In this figure the 25% contour of the probability density for the systems of particles with WCA interactions is shown for three temperatures (data sets D, E, G). As expected, these results are qualitatively comparable to those shown in Sec. 3.2.1. The same trends can be observed in Figs. 3.10(b) and 3.10(c) for the same data sets as in Fig. 3.10(a) but for different values of the global correlation $C(t_3, t_1)$. 
Figure 3.9: 2D contours of constant joint probability density $\rho(|\Delta \sigma_r|, |\Delta \pi_r|)$, computed using coarse graining boxes containing 125 particles on average. By subtracting the global quantities from the local quantities we avoid trivial effects due to differences in the global values. Each panel from A to H contains results from the corresponding data set, for $C(t_1, t_3) \approx 0.23$, with the times chosen as late as possible within each data set. Each set of three concentric contours is chosen so that they enclose 25%, 50% and 75% of the total probability.
Figure 3.10: Comparison of the contours enclosing 25% of the probability for the aging WCA systems at three different temperatures (D, E and G). The three different panels correspond to different values of $C(t_3, t_1)$. 
Furthermore, one can also analyze the effects of choosing different conditions from the ones chosen in Fig. 3.9, for instance, by comparing the results shown in Fig. 3.9 with results obtained for smaller coarse graining regions or for shorter times in the aging regime. This comparison is shown in Fig. 3.11, where the 25% probability contours for data set B are shown for three conditions. The contour labeled B is the one shown already in Fig. 3.9. The contour labeled B’ corresponds to the same time, but with coarse graining regions containing on average 23 particles instead of 125. This leads to less averaging and stronger fluctuations, but also, since fluctuations correlated over shorter distances are no longer preferentially suppressed, the shape of the contour is no longer dominated by collective modes, and thus contour B’ extends more in the direction of $|\Delta \sigma_\vec{r}|$ than contour B. The contour labeled B” corresponds to the same coarse graining size of contour B, but

![Figure 3.11: 25% probability contours for data set B with $C(t_1, t_3) \approx 0.23$. Contour B corresponds to the 25% of the probability density shown in Fig. 3.9. Contour B’ corresponds to the same times as contour B, but with a much smaller coarse graining size. Contour label B” corresponds to the same coarse graining size as B, but with much shorter times.](image-url)
with much shorter times. This leads to stronger finite time effects, analogous to the ones found at slightly higher temperatures, and as expected the contour is less anisotropic, and indeed, it resembles the contours corresponding to $T/T_c \approx 1.1$.

### 3.2.3 Measuring the Transverse and Longitudinal Fluctuations

A more extensive analysis of the fluctuations can be performed by separating longitudinal and transverse components in Eqs. (3.20) and (3.21).

First, recall that $\sigma$ is a purely longitudinal quantity (see Eq. (3.19)), therefore $\sigma_{T} = 0$ and $\Phi_{31,\vec{r}} = \Phi_{21,\vec{r}} + \Phi_{32,\vec{r}}$. Now, the following two assumptions are made. One is that the transverse and longitudinal fluctuations are not correlated to each other. The other is that all slow fluctuations are transverse, or in other words, that longitudinal fluctuations are short range correlated in time, or at least they are correlated over times that are shorter than the shortest time interval between the configurations that are being considered. This leads to the conditions

\[
\left\langle \delta \Phi_{ab,\vec{r}} \Phi_{cd,\vec{r}'} \right\rangle = 0 \quad \forall \quad a, b, c, d, \vec{r}, \vec{r}'
\]

(3.23)

and

\[
\left\langle \delta \Phi_{ab,\vec{r}} \Phi_{cd,\vec{r}'} \right\rangle = 0 \quad \text{for} \quad a \neq c \quad \text{or} \quad b \neq d, \quad \forall \quad \vec{r}, \vec{r}'.
\]

(3.24)

Here $\delta x \equiv x - \langle x \rangle$. By using Eq. (3.17), it can be shown that the transverse components of Eqs. (3.20) and (3.21) are given by

\[
\pi_{1,\vec{r}} = \frac{1}{\sqrt{2}}(\Phi_{21,\vec{r}} - \Phi_{32,\vec{r}})
\]

(3.25)

and

\[
\pi_{2,\vec{r}} = \frac{3}{\sqrt{6}}(\Phi_{21,\vec{r}} + \Phi_{32,\vec{r}}).
\]

(3.26)

Regarding the longitudinal components, by using Eq. (3.24) it can be shown that
\[
\langle (\delta \sigma_r) \rangle^2 = \frac{1}{3} \left[ \langle (\delta \Phi_{21})^2 \rangle + \langle (\delta \Phi_{32})^2 \rangle + \langle (\delta \Phi_{31})^2 \rangle \right], \tag{3.27}
\]

\[
\langle (\delta \pi_1) \rangle^2 = \frac{1}{2} \left[ \langle (\delta \Phi_{21})^2 \rangle + \langle (\delta \Phi_{32})^2 \rangle \right], \tag{3.28}
\]

and

\[
\langle (\delta \pi_2) \rangle^2 = \frac{1}{6} \left[ \langle (\delta \Phi_{21})^2 \rangle + \langle (\delta \Phi_{32})^2 \rangle + 4 \langle (\delta \Phi_{31})^2 \rangle \right]. \tag{3.29}
\]

From the last three equations it can be easily seen that

\[
\langle (\delta \pi_1)^2 + (\delta \pi_2)^2 - 2(\delta \sigma)^2 \rangle = 0. \tag{3.30}
\]

By using Eq. (3.23), the variance of the transverse fluctuations can be computed by combining the fluctuations of Eqs. (3.19), (3.20) and (3.21) in the following way

\[
\langle (\delta \pi_1)^T + (\delta \pi_2)^T \rangle = \langle (\delta \pi_1)^2 + (\delta \pi_2)^2 - 2(\delta \sigma)^2 \rangle. \tag{3.31}
\]

Further, the correlation volumes (in units of the coarse graining volume \( V_{cg} \)) can be estimated by using the formula

\[
V_{corr} \equiv \chi_{4,a} \left[ \left| \langle (\delta a)_r \rangle^2 \right| \right], \tag{3.32}
\]

where \( \chi_{4,a} \equiv V \langle (\delta \bar{a})^2 \rangle \), \( a_r \) is a local coarse grained variable, \( \bar{a} \equiv \frac{1}{L^d} \int a_r \) is the spatial average of \( a_r \), and \( V = L^d \) is the volume of the system. Therefore, by using these equations together with Eq. (3.31) one can estimate the correlation volume of transverse and longitudinal fluctuations, respectively given by

\[
V_T = \frac{\chi_{4,\pi_1} + \chi_{4,\pi_2}}{V_{cg} \langle (\delta \pi_1)^T + (\delta \pi_2)^T \rangle} = \frac{V \langle (\delta \pi_1)^2 + (\delta \pi_2)^2 - 2(\delta \sigma)^2 \rangle}{V_{cg} \langle (\delta \pi_1)^2 + (\delta \pi_2)^2 - 2(\delta \sigma)^2 \rangle}, \tag{3.33}
\]
If the time reparametrization hypothesis is correct, it is expected that the variance as well as the correlation volume of the transverse fluctuations will grow together with those corresponding to the dynamical heterogeneities. It is also expected that the variance and correlation volume of the longitudinal fluctuations should be insensitive to changes in the variance and correlation volume of the dynamical heterogeneities.

The top panel of Fig. 3.12 shows the ratio between the variances of local transverse fluctuations and longitudinal fluctuations, 
\[ \frac{\langle (\delta \pi_1)^2 + (\delta \pi_2)^2 \rangle}{\langle (\delta \sigma)^2 \rangle} \] ("variance anisotropy ratio") (see Eq. (3.31)), as a function of the initial time \( t_1 \). Similarly, in the bottom panel of Fig. 3.12, the ratio between the correlation volumes of transverse and longitudinal fluctuations, \( V^T/V^L \) ("correlation volume anisotropy ratio") (see Eqs. (3.33) and (3.34)), is plotted also as a function of \( t_1 \). It can be observed that for aging systems both ratios grow as \( t_1 \) increases, as one could expect from the fact that at later times the reparametrizations symmetry breaking terms in the action should become progressively weaker [32]. For the equilibrium data sets, the dynamics is time translation invariant (TTI), and we observe, as expected, that both anisotropies are independent of \( t_1 \).

Moreover, Fig. 3.13 shows the same two ratios as functions of the strength of the dynamical heterogeneities, measured by the dynamical susceptibility
\[ \chi_4(t_3, t_1) \equiv \chi_{4,C_{31}} = \mathcal{N} \left[ \left( \langle C_{31}^2 \rangle - \langle C_{31} \rangle^2 \right) \right] \].

It can be seen that both anisotropy ratios grow when \( \chi_{4,C_{31}} \) increases, i.e., as the dynamical heterogeneity becomes more pronounced. Although the same qualitative behavior is observed for all data sets, the curves are different for different systems and temperatures.

The results presented in Fig. 3.13 correspond to the value \( C(t_3, t_1) \approx 0.23 \), but similar results can be obtained for different values of \( C(t_3, t_1) \), as one can expect from Fig. 3.10. Results for three different values of \( C(t_3, t_1) \) are shown in Fig. 3.14, where the variance
Figure 3.12: *Top panel:* Ratio between the variances of transverse and longitudinal fluctuations as a function of the initial time $t_1$, for $C(t_3,t_1) \approx 0.23$. *Bottom panel:* Ratio between the correlation volumes of transverse and longitudinal fluctuations as a function of $t_1$, for the value $C(t_3,t_1) \approx 0.23$. 
Figure 3.13: Top panel: Ratio between the variances of transverse and longitudinal fluctuations as a function of the strength of the dynamical heterogeneity, measured by $\chi_{4,C_{31}} \equiv \chi_4(t_3,t_1)$, for the value $C(t_3,t_1) \approx 0.23$. Bottom panel: Ratio between the correlation volumes of transverse and longitudinal fluctuations as a function of $\chi_{4,C_{31}}$, for $C(t_3,t_1) \approx 0.23$. 
Figure 3.14: Ratio between the variances of transverse and longitudinal fluctuations as a function of $\chi_{4,c_{31}}$, for systems B and D for the values $C(t_3, t_1) \approx 0.23, 0.33, \text{and } 0.44$.

The variance anisotropy ratio for data sets B and D is plotted as a function of $\chi_{4,c_{31}}$ for the values $C(t_3, t_1) \approx 0.23, 0.33 \text{ and } 0.44$. This figure shows that the variance anisotropy ratio grows with the strength of the dynamical heterogeneity, for all three fixed values of $C(t_3, t_1)$.

According to the hypothesis, dynamical heterogeneity originates in the Goldstone modes associated to fluctuations in the time reparametrization (see Eq. (3.15)). Therefore, the hypothesis implies that the correlation volume of the dynamical heterogeneity should be similar to the correlation volumes of the transverse components of the variables $\pi_1$ and $\pi_2$, and the longitudinal variable $\sigma$ should be less correlated in space. The results, shown in Fig. 3.15, show that this is indeed the case. The correlation volume corresponding to the transverse fluctuations, $V^T$, closely tracks the one corresponding to the dynamical heterogeneities, $V_{c_{31}}$, and they both grow together as the temperature is reduced or the
timescale is increased. By contrast, for the longitudinal fluctuations $\sigma$ I find that their correlation volume $V^L$ is small and essentially constant; it does not correlate with the correlation volume of the dynamical heterogeneity, nor with the temperature or the time scale. In fact, despite the large error bars and the presence of some outlier points, the figure shows a partial data collapse between different systems, both for the case of transverse and for the case of longitudinal correlation volumes. In the case of longitudinal fluctuations, this may be a trivial effect due to the correlation volumes being smaller than the volume of the coarse graining regions used to define the variables. In the case of the transverse fluctuations, the correlation volumes go well beyond the volume of the coarse graining regions, and the partial collapse in the results might be evidence of some sort of universality, but more work will be needed in order to decide this question one way or another.

### 3.3 Discussion

In this project, I tested in structural glass simulated data the hypothesis that dynamical heterogeneity arises from Goldstone modes related to a broken continuous symmetry under time reparametrizations. In other words, I tested whether dynamical heterogeneity is associated with the presence of spatially correlated fluctuations in the time variables.

In these tests, the triangular relations were used in two ways. In the first way, the global curve that probes the hypothesis was constructed in a similar way as in Ref. [33]. In the second way, the global curve was constructed such that the 2d joint probability distributions allow one to distinguish between time reparametrization fluctuations (transverse fluctuations) and other fluctuations (longitudinal fluctuations). Also, I applied a test for possible correlations of both the strength and the correlation volume of the fluctuations in the time variable with the dynamical susceptibility $\chi_4$, which is normally
used to probe dynamical heterogeneity. Altogether, I found that at the lowest temperatures, for the longest timescales and for the largest coarse graining lengths, the transverse fluctuations became stronger than the longitudinal fluctuations, which is consistent with the hypothesis. This was the case independently of the system considered. Moreover, I found that the correlation volumes of the time reparametrization fluctuations were proportional to the correlation volumes of the dynamical heterogeneity, while the correlation volumes of the longitudinal fluctuations were small and independent of the correlation volumes of the dynamical heterogeneity. All of this was despite the fact that, to simplify the analysis, some extra conditions on the form of the correlations were
imposed, which may have made the agreement with the hypothesis appear less good than
it would have been otherwise.

With respect to universality, the evidence is mixed. On the one hand, there were clear
differences in the details of the results for different systems, for example for the anisotropy
ratios. On the other hand, all the trends observed in this work were the same across
systems, and the results for the correlation volumes did show some hints of universality,
although the relatively large noise in this measurement did not allow for definite
conclusions to be drawn. In any case, the commonality in the results is strong enough to
suggest that other systems may display similar qualitative behaviors. Thus, it is expected
that it would be very instructive to apply the same kind of test to data from other slowly
relaxing systems, such as particle tracking data from glassy colloids [17, 18, 22, 23] and
from granular systems close to jamming [25, 41].

Finally, considering the success of the tests presented here, it becomes natural to ask
if it is possible to extract from the data the actual fluctuating reparametrization \( \varphi_\tau(t) \), and
to study its properties directly. In fact, Ref. [40] already shows some progress in that
direction.
4 Conclusions

In this work, I studied dynamical heterogeneity in granular fluids and in structural glasses.

In granular fluids, I found that, qualitatively, the growing behavior of the dynamical regions shows similar trends as in other glassy systems. However, the highest values of the correlation length $\xi$ found in this work are larger than in any previous result obtained either from experiments or simulations. Both the dynamical susceptibility $\chi_4(t)$ and the correlation length $\xi(t)$ as functions of the packing fraction $\phi$ can be fitted by power law divergences as predicted by mode coupling theory. However, the relaxation time $\tau_\alpha$ as a function of $\phi$ was better fit to an exponential form than to a power law. Also, I found that the four point correlation function $S_4(q, \tau_\alpha)$ is well described by an Ornstein-Zernike fit $S_4(q, \tau_\alpha) \approx \chi_4(\tau_\alpha)/(1 + (q\xi(\tau_\alpha))^2)$ in the limit of small values of $q$. This fit does not describe the data accurately for $q\xi > 3$. However, the scaling relation $S_4(q, \tau_\alpha)/\chi_4(\tau_\alpha) = s[q\xi(\tau_\alpha)]$ is a good approximation over the whole $q\xi$ range (for $q < 0.3$). Additionally, the exponent $d - p \approx 1.63$ found from the relationship $\chi_4(\tau_\alpha) \approx A(\xi(\tau_\alpha))^{d-p}$ has approximately the same value for different choices of the cutoff parameter $a$ used to define the observables and for different values of the coefficient of restitution $\varepsilon$. This suggests that the correlated clusters have the same fractal dimension independently of the choice of $a$ and $\varepsilon$.

In the structural glass, I found the results to be consistent with the time reparametrization invariance hypothesis. This is that for low temperatures, long timescales, and large coarse graining lengths, the fluctuations related to the time reparametrization symmetry (transverse fluctuations) became stronger than other fluctuations (longitudinal fluctuations). Similar qualitative results were obtained for different glass forming systems. Also, the correlation volumes of the time reparametrization fluctuations were found to be proportional to the correlation volumes of
the dynamical heterogeneity, while the correlation volumes of fluctuations unrelated to the
time reparametrization symmetry were found to be small and independent of the
correlation volumes of the dynamical heterogeneity. This proportionality seems to hold
for all systems studied, but due to the relatively large noise in this measurement it is not
possible to conclude if our results are universal.

In general, this work provides evidence that indicates that dynamical heterogeneity
for granular systems can be described in a similar way as for other glassy systems, and
that many properties of dynamical heterogeneity in glass models could be explained as a
consequence of a continuous broken time reparametrization symmetry.

The results of this work suggest other questions to ask, such as: Could the properties
of the dynamical heterogeneity in a granular system be described by the time
reparametrization fluctuation hypothesis as it was shown here for structural glasses? Can
the power law behaviors of $\chi_4(\tau_\alpha)$ and $\xi(\tau_\alpha)$ vs $\phi$ be made compatible with the exponential
behavior of $\tau_\alpha$ vs $\phi$? If not, should other fitting forms be used? Is the fractal dimension of
the clusters changing for times much longer than the relaxation time? How is the
correlation length growing for larger values of $a$ than the ones used here? What is the
relationship between timescales and length scales for packing fractions closer to the
jamming transition? Is there a functional form that describes the scaling relationship
$S_4(q, \tau_\alpha)/\chi_4(\tau_\alpha) = s[q\xi(\tau_\alpha)]$ better than the Ornstein-Zernike form? Is the geometry of the
clusters in structural glasses also independent of the choice of the cutoff length $a$ as in the
case investigated here for granular fluids? Can a correlation length be obtained for the
time reparametrization fluctuations in glasses? Can the relaxation mechanism of the
correlated regions that lead to a non-exponential relaxation in glasses be inferred by
studying the properties of the time reparametrization fluctuations?
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