Micromechanics and rheology of hard and soft-sphere colloidal glasses
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6. Visualization of incipient deformation by indenting colloidal glasses

In chapter 4 we investigated the yielding behavior of hard and soft suspensions via their macroscopic rheology, while in chapter 5 we elucidated the microscopic relaxation of hard and soft-sphere suspensions. In both cases, a central question concerns incipient plasticity – the onset of permanent deformation – that is central to their relaxation, aging, and yielding. In this chapter we elucidate the onset of permanent deformation using indentation. By concentrating the stress in small volumes, we initiate plastic deformation in a controlled way, and we investigate on the single particle level how the material starts to yield. We observe a surprising hierarchical structure of incipient deformation that indicates a critical behaviour of the glass upon yielding.

6.1: Introduction

Incipient plasticity addresses a crucial step in the deformation of materials. It marks the onset of permanent deformation due to irreversible atomic rearrangements. At small applied strain, solids deform elastically, being able to recover their shape when the strain is removed. However, at larger applied strain, irreversible rearrangements occur, leading to permanent deformation. This onset of permanent deformation reflects a symmetry change from time reversible to irreversible atomic motion. Glasses are frozen on experimental time scales, and the onset of irreversible rearrangements is crucial for the understanding of their relaxation, aging and deformation. Nanometre-scale indentation experiments can provide insight into the incipient plasticity, being able to resolve the very earliest stages of deformation [1-3]. While nanoindentation experiments on atomic glasses allow the resolution of individual elementary plastic events from force-displacement measurements [4, 5], the direct imaging of elementary relaxation, and the microscopic displacements at the onset of plastic deformation are extremely difficult to observe directly in atomic glasses. In this chapter, we use an analogue of nano-indentation performed on a colloidal glass to obtain direct images of the incipient plasticity, allowing us to elucidate the onset of permanent deformation. We visualize the microscopic strain directly by following distorted nearest neighbour configurations, and observe a surprising...
hierarchical structure: at the onset of irreversible deformation, the strain acquires a robust fractal structure, similar to the structure of solid networks in the irreversible aggregation of particles, and we measure the fractal dimension directly from the imaged strain distribution. These results give direct evidence that the onset of permanent deformation has the hallmarks of a critical point, in agreement with recent theoretical work on fibrous networks [6]. This critical behavior is of crucial importance for the application of amorphous materials as new engineering materials at small scales [7].

6.2: The mechanics of elastic contact: Hertz’s theory

In 1882, Hertz analysed the stress at the contact of two elastic solid bodies [8] motivated by his study about Newton’s optical interference fringes in the gap between two glass lenses. He was concerned about the influence of elastic deformation of the lens surfaces during contact due to the pressure between them. He started to investigate elasticity in order to understand the reversible change in the lens surface change. In his theory, Hertz made the following assumptions: The surfaces are continuous and non-conforming; the strains are small; each body can be considered as an elastic half-space; the contact is frictionless [9].

The fundamental analysis of these problems in the elastic regime was first given by Hertz (1881); and then worked out further by Huber (1904), by Fuchs (1913), and by Morton & Close (1992) for the case where the area of contact is a circle, whilst Thomas & Hoersch (1930) have given a more general solution for the case of non-spherical contact area.

The theory provides analytic solutions of the stresses and strains produced when a spherical indenter is pressed against the plane surface of a semi-infinite elastic body. The
surface of contact is bounded by a circle—the circle of contact. When a sphere of radius \( R \) is pressed with a total pressure \( P \) against the plane surface with radius of \( a \) (Fig. 6.1), then by a reduction of Hertz’s equations as given by Love, it can be shown that the surface of the circle of contact is

\[
a = \frac{3P}{16 \rho}, \quad \text{where} \quad (6.1)
\]

\[
\vartheta = \frac{4(1-\mu^2)}{E} \quad (6.2)
\]

Here \( E \) and \( \mu \) are respectively, the Young’s modulus and Poisson’s ratio of the material and \( \rho=1/R \). In Hertz’s solution, the displacement of a point on the surface within the area of contact is given by

\[
u = \frac{1}{2} \left[ r^2 + Z^2 - a^2 \pm \sqrt{(r^2 + Z^2 - a^2)^2 + 4a^2Z^2} \right]. \quad (6.3)
\]

where \( r \) and \( Z \) are the distances from the origin (contact point) in cylinder coordinates. From the displacement, we can calculate strain and stress components in the indented elastic body [10]. The derivation is lengthy, and we give only the result here. The normal stresses inside the elastic body are:

\[
\sigma_{xx} = p' \left\{ \frac{1 - 2\mu a^2}{3 r^2} \left[ 1 - \left( \frac{Z}{\sqrt{u}} \right)^3 \right] + \frac{Z}{\sqrt{u}} \left[ 2\mu + \frac{(1 + \mu)u}{a^2 + u} - (1 + \mu) \frac{\sqrt{u}}{a} \arctg \frac{a}{\sqrt{u}} \right] \right\} \quad (6.4)
\]

\[
\sigma_{yy} = -p' \left\{ \frac{1 - 2\mu a^2}{3 r^2} \left[ 1 - \left( \frac{z}{\sqrt{u}} \right)^3 \right] + \left( \frac{Z}{\sqrt{u}} \right)^3 \frac{a^2u}{u^2 + a^2Z^2} + \frac{Z}{\sqrt{u}} \left[ \frac{(1 - \mu)u}{a^2 + u} + (1 + \mu) \frac{\sqrt{u}}{a} \arctg \frac{a}{\sqrt{u}} - 2 \right] \right\} \quad (6.5)
\]

\[
\sigma_{zz} = p' \left( \frac{Z}{\sqrt{u}} \right)^3 \frac{a^2u}{u^2 + a^2Z^2} \quad (6.6)
\]

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While the shear stress in the indentation plane is

\[ \sigma_{yz} = p' \frac{y z^2}{u^2 + a^2 z^2} \frac{a^2 \sqrt{u}}{a^2 + u}. \]  

(6.7)

Here \( p' = \frac{3p}{16 \pi a^2} \) [11].

In our glass, deformation is driven by the maximum shear stress: It is possible to find three orthogonal planes such that shear stress vanishes; these planes are called the principal planes and the normal stresses on these planes are called the principal stresses. The principal stresses are the maximum (\( \sigma_1 \)) and minimum (\( \sigma_2 \)) possible values of the normal stresses. The maximum shear stress is found by taking the maximum difference between the principal stresses:

\[ \sigma_{max} = \frac{|\sigma_1 - \sigma_2|}{2} \]  

(6.8)

a distribution of maximum shear stress within an elastic continuum is given in Fig. 6.2 [12].

**Figure 6.2:** Contour plot showing the distribution of the maximum shear stress in a continuous linear-elastic medium. The peak value of shear stress, \( \sigma_{max} \) occurs directly below the center of the spherical indenter at a distance of about half the contact radius (a) below the specimen surface. \( P_m \) is the mean contact pressure [13].
For an isotropic linear elastic material, stresses and strains are related via:

\[
\begin{pmatrix}
\varepsilon_{xx} \\
\varepsilon_{yy} \\
\varepsilon_{zz} \\
\varepsilon_{xy} \\
\varepsilon_{xz} \\
\varepsilon_{yz}
\end{pmatrix} = \frac{1}{E} \begin{pmatrix}
1 & -\mu & -\mu & 0 & 0 & 0 \\
-\mu & 1 & -\mu & 0 & 0 & 0 \\
-\mu & -\mu & 1 & 0 & 0 & 0 \\
0 & 0 & 0 & 2(1+\mu) & 0 & 0 \\
0 & 0 & 0 & 0 & 2(1+\mu) & 0 \\
0 & 0 & 0 & 0 & 0 & 2(1+\mu)
\end{pmatrix} \times \begin{pmatrix}
\sigma_{xx} \\
\sigma_{yy} \\
\sigma_{zz} \\
\sigma_{xy} \\
\sigma_{xz} \\
\sigma_{yz}
\end{pmatrix}
\] (6.9),

where \( \varepsilon_{ij} \) are shear strain components and \( \varepsilon_{kk} \) are the normal strains, \( \mu \) is the Poisson ratio and \( E \) is young modulus. The total energy associated with the strain components is:

\[
E_{el} = V G \left( \varepsilon_{ij}^2 + \frac{\mu}{1-2\mu} \varepsilon_{kk}^2 \right) \tag{6.10}
\]

Here \( V \) is the volume element of the strain and \( G \) is the shear modulus which is equal to \( E/(2(1+\mu)) \). Because in our analysis, we use the nearest neighbours for the calculation of strain, we take \( V = 4/3 \pi R_0^3 \), where \( R_0 = r_{\text{particle}} \), \( r_{\text{particle}} \) is the minimum of the pair correlation function, see Fig 6.3.

**6.3: Experimental setup**

To investigate the indentation of a glass with single-particle resolution we use a colloidal glass and confocal microscopy to track the individual particles. We prepare a colloidal glass by sedimenting 1.55 \( \mu \)m diameter silica particles onto a cover slip by centrifugation. The particles are suspended in a mixture of water and dimethylsulfoxide (DMSO) with volume fractions of 1/3 and 2/3, respectively, that matches their refractive index. The cover slip surface is coated with a 5\( \mu \)m thick layer of polydisperse colloidal particles (size distribution 1-5\( \mu \)m) to avoid boundary-induced crystallization. The resulting colloidal amorphous film has a volume fraction of \( \phi \sim 0.60 \), well inside the glassy state [14]. The structure of our glass is characterized by the pair correlation function while we show in Fig. 6.3. A sewing needle attached to a piezoelectric translation stage is used to indent the colloidal glass. The needle has an almost hemispherical tip with radius \( \sim 38 \mu \)m; the length scale ratios of the particle size, the film thickness and the tip diameter are
similar to those used in nanoindentation experiments on conventional (atomic) materials. The needle touches the surface of the glass at time $t=0$, when we start recording. A piezo is used to lower the needle very accurately with a speed of $2.9 \, \mu m/h$ into the colloidal glass. We use confocal microscopy to image the motion of individual particles in the glass. Every single minute, a three-dimensional image of a volume $66 \, \mu m \times 66 \, \mu m \times 48 \, \mu m$ below the tip is acquired (Fig. 6.5a). This technique allows us to follow the incipient plastic deformation directly at the particle scale. We determine individual particle positions in three dimensions with an accuracy of $0.3 \, \mu m$ in the horizontal, and $0.5 \, \mu m$ in the vertical direction [15]. We start recording images 15 minutes before starting the indentation. We then acquire images every 60 sec over a time interval of 25 min from indented glass to track the motion of $\sim 3 \times 10^4$ particles in the imaged section. One image needs 45 seconds to acquire. As a reference before the indentation starts, we also acquire image stacks of the quiescent glass. This allows us to determine the elastic properties of the amorphous film from thermally induced fluctuation of particle positions.

![Figure 6.3: Radial pair correlation function](image)

The pair correlation function shows the structure of the colloidal glass. The first minimum, $r_0$ (vertical dashed line), indicates the boundary of the nearest-neighbour shell.

6.4: Results - quiescent glass

We use thermally induced strain fluctuations in the quiescent glass to determine the elastic modulus of the amorphous film. Due to thermal fluctuations, oscillations of
strain will occur spontaneously. In thermal equilibrium, the probability of excitation of elastic strain energy $E_{el}$ is $P \propto e^{-\frac{E_{el}}{kT}}$. Replacing $E_{el}$ according to Eqn. (6.10), we obtain

$$P \propto \exp\left(-\frac{VG\left(\varepsilon_{ij}^2 + \frac{\mu}{1-2\mu}\varepsilon_{kk}^2\right)}{k_b T}\right)$$  \hspace{1cm} (6.11)

Therefore, by plotting the relative frequency of strain energies as a function of strain energy magnitude we can determine the elastic modulus from the slope of the data. We plot $P(E_{el})$ as a function of $E_{el}/VG$ for all particles in the volume in Fig 6.4. The data can indeed reasonably well be fitted with a line, meaning that the Boltzmann distribution describes the observed strain distribution reasonably well. By linear extrapolation, we obtain the shear modulus $G=0.189 \text{ Pa}$.

![Figure 6.4: Probability distribution of elastic energy.](image)

6.5: Results- indented glass

6.5.1: Particle displacements and strain distribution

We show particle displacements after 21 min of indentation in Fig. 6.5b. Large displacements concentrate below the tip, as expected. We find that, however, even far away from the tip, pronounced displacements occur, something that is not expected for a material that behaves as a continuum elastic material.
Figure 6.5: Indentation setup and strain distribution

(a) Schematic showing the needle tip with respect to the amorphous colloidal film, and the imaged 66 μm by 66 μm by 30 μm volume. (b-d) Particle displacements and strain distribution after 23 min of indentation. (b) Particle displacements in a 5 μm thick section centered below the needle. Arrows are enlarged by a factor of two for clarity. (c, d) 66 μm by 66 μm by 25 μm reconstruction of the strain distribution. Particle color indicates the magnitude of the maximum local shear strain, $\varepsilon_{\text{max}}$ (see color bar). In (d), particles with $\varepsilon_{\text{max}} < 0.1$ have been omitted for clarity. (e) shows the maximum shear strain as a function of height below the tip, experimental measurement (red dots) and continuum elasticity prediction (dashed curve). (f)-(h) Time series showing the evolution of the maximum shear strain in a 66 μm by 66 μm by 5 μm section, 10 μm below the needle, after 9 min (f), 15 min (g), and 21 min (h) of indentation.
To explore the deformation of the glass in more detail, we determine the local strain from distorted nearest neighbour configurations. As described in sec. 3.8 a good measure of the local shear deformation is the maximum shear strain, $\varepsilon_{\text{max}} = |\varepsilon_1 - \varepsilon_2|/2$ [16], an invariant of the strain tensor, where, $\varepsilon_1$ and $\varepsilon_2$ are the largest and smallest eigenvalues of the strain tensor. The maximum shear strain $\varepsilon_{\text{max}}$ reflects the shear strain acting along the principal axes of the strain tensor, and has been used as a good measure of strain in the inhomogeneous deformation of isotropic materials [16]. To visualize its distribution, for each particle, we determine values of $\varepsilon_{\text{max}}$ from the eigenvalues of the strain tensor associated with the particle. Colour representations of the resulting strain distributions are shown in Fig. 6.5c and d. Orange and yellow spheres indicate particles with large $\varepsilon_{\text{max}}$, while blue particles indicate small $\varepsilon_{\text{max}}$. In Fig. 6.5d, we have omitted particles with $\varepsilon_{\text{max}} < 0.11$ for clarity. Orange particles accumulate below the tip, indicating high local shear strain. A strain maximum occurs roughly 10 $\mu$m below the tip (Fig. 6.5e), in reasonable agreement with continuum theory predictions. In contrast to continuum elasticity predictions, however, large shear strain occurs surprisingly far away from the tip. To demonstrate this, we focus on a horizontal section, 10 $\mu$m below the tip, and show the time evolution of strain in Figs. 6.5f to h. Already at early times (Fig. 6.5f and g), large shear strain is observed throughout the field of view as indicated by the occurrence of yellow spheres far away from the tip. This strain distribution shows a surprising cascade structure as demonstrated in Fig. 6.6a. Yellow zones concentrate at preferred distances to the origin (dashed circles), indicating a cascade-like structure of deformation. Similar strain patterns are observed in all components of strain. As an example, we show the distribution of normal strain $\varepsilon_{zz}$ in Fig. 6.6b. Blue spheres in the centre reflect directly the compressive strain exerted by the tip, while the pattern of blue and red spheres indicate the occurrence of compression and dilation at positions that coincide with those of maximum shear strain (Fig. 6.6a).
Figure 6.6: Cascade structure of the incipient deformation

(a, b) Strain distribution in a 5 μm thick section 10μm below the tip after 21min of indentation. a, Distribution of the maximum shear strain, $\varepsilon_{\text{max}}$. b, Distribution of normal strain $\varepsilon_{zz}$. (c) Angle-averaged strain as a function of distance from the center of the tip. Symbols indicate the measured values of $\varepsilon_{\text{max}}$ (red dots), $\varepsilon_{zz}$ (green dots), and the radial shear strain $\varepsilon_{rz}$ (violet dots). Dashed lines indicate continuum predictions for $\varepsilon_{\text{max}}$, $\varepsilon_{zz}$, and $\varepsilon_{rz}$, respectively. Arrows indicate pronounced deviations from the predicted elastic distribution associated with the onset of permanent deformation.

6.5.2: Comparing strain value to the elastic strain distribution predicted by Hertzian theory.

The cascade structure is a characteristic property of the emergent plastic deformation; to show this, we compare the observed strain distribution with the elastic strain distribution predicted by continuum elasticity. As shown in 6.2 the elastic strain
distribution depends only on the radius of contact, \( a \), between the indenter and the medium, and the maximum normal pressure \( P \) in the centre of contact. To determine the elastic strain distribution corresponding to Figs. 6.6a and b, we take advantage of our direct imaging, and measure the contact radius directly from the imaged contact surface between the tip and the colloidal glass, obtaining \( a = 22 \, \mu m \). We then adjust \( P \) in order to best fit the experimental strain distributions. We obtain \( P = 0.028 \, (pa) \) correspond to a normal strain of \( \varepsilon_n = 0.083 \). The resultant elastic strain distributions are shown in Fig. 6.6c (dashed lines). Also shown by dots are the measured strain distributions. Striking differences occur at \( R_1 = 10 \), \( R_2 = 25 \) and \( R_3 = 38 \, \mu m \) (arrows) in all components of strain. These distances coincide precisely with the location of the circles in Fig. 6.6a and b; we associate these deviations from the elastic strain distribution with the onset of plastic relaxation.

**6.5.3: Time evolution of strain**

Load-displacement curves of metallic glasses exhibit characteristic bursts at the onset of plastic flow [1] that have been associated with the nucleation of shear bands, where slip occurs collectively along a surface. Such shear bands occur at an intermediate regime of indentation rates where the glass exhibits inhomogeneous flow [5]. The long time scale of our colloidal glass permits us to access the regime of low indentation rates, where uniform flow occurs [17]: Surprisingly, we observe a cascade structure of the incipient plastic deformation, characterized by strong spatial correlation. The excellent time resolution of our technique allows us to even follow the propagation of the incipient plastic deformation in time: We focus on particles with large shear strain, and follow their time evolution in Fig. 6.7a-c. The incipient plastic deformation penetrates the glass with a characteristic propagation time. To determine this time scale, we measure the delay time of plastic activity at the distances \( R_1 \), \( R_2 \) and \( R_3 \) corresponding to the circles in Fig. 6.6a and b. We correlate the angle-averaged strain \( \varepsilon_i(t) \) at \( R_i \) at time \( t \) with the strain \( \varepsilon_j(t + \Delta t) \) at \( R_j \) at some later time, \( t + \Delta t \). The corresponding correlation functions, shown in Fig. 6.7d, reveal distinct time correlation: Maxima at \( \Delta t = 6 \), 8 and 14 min demarcate characteristic delay times associated with the propagation of plastic deformation, and reflect the time
necessary to nucleate new plastic zones in the strain field of existing ones. Thus we conclude that in addition to the spatial correlation, there is also strong time correlation in the incipient plastic deformation.

**Figure 6.7: Time evolution of strain**

(a)-(c) Time series showing the evolution of regions of high strain after 16, 20, and 24 min of indentation. (d) Time correlation of strain between the inner and middle (blue), middle and outer (red) and middle and outer (green) dashed circle in Figs. (a-c). The time correlation is calculated by correlating the average strain within cylindrical shells of thickness 4 μm, and radii \( R_1 = 10, R_2 = 25, \) and \( R_3 = 38 \) μm, centered at the origin. Maxima indicate the characteristic delay times of plasticity, \( \Delta t_{12} = 6, \Delta t_{23} = 8 \) and \( \Delta t_{13} = 14 \) min.

**6.5.4: Fractal deformation field**

This strong spatial and temporal correlation results in a complex, highly correlated network of deformation. To elucidate it, we explore the analogy with fractal structures encountered in colloidal particle aggregation, where strong interparticle attraction leads to open, hierarchical structures characterized by a robust fractal dimension [20]. While for space-filling structures, the number \( N(r) \) of particles within a distance \( r \) grows as \( N(r) \propto r^D \), where \( D \) is the dimension of space, in open, fractal structures, \( N(r) \propto r^{d_f} \), where \( d_f < D \) is the fractal dimension. Surprisingly, we find that a similar description applies to the distribution of strain during the incipient deformation. To show this, we again focus on high strain particles, and determine their number \( N(r) \) within a lateral distance \( r \) from the origin. We plot \( N \) as a function of \( r \) in Fig. 6.8a, inset. Within the highly deformed centre, \( N \sim r^2 \), indicating a compact space-filling structure. However, outside the central zone, \( N \) grows slower, with a slope of \( \sim 1.2 \) indicating a fractal structure of deformation. To show this most clearly, we normalize by the homogeneous
distribution, $\bar{N} = N/\pi \bar{\rho} r^2$, where $\bar{\rho}$ is the average density of high-strain particles, and show the normalized number of particles in Fig. 6.8a, main panel. The change from a horizontal to a negative slope of the data indicates the transition from a compact to a fractal structure of deformation. The excellent collapse of the data taken after different times of indentation demonstrates that this fractal structure is a robust property of the incipient deformation of the glass.

**Figure 6.8: Fractal deformation field**

(a) Scaling of the number $\bar{N}$ of high-strain particles, normalized by the homogeneous distribution, as a function of distance $r$ from the origin. Two regimes with different scaling are delineated by the vertical dashed line. For $r<15 \ \mu m$, $\bar{N}$ is constant, indicating a compact, space-filling structure, while for $r>15 \ \mu m$ $\bar{N} \sim r^{-1.2}$ indicating a fractal structure. Colors indicate increasing time intervals of indentation, $t=15$ (red), 16 (violet), 17 (gray), 18 (green), 19 (blue), 20 (yellow) and 24 min (turquoise). Inset: Same data without normalization shows the fractal scaling of the number of high-strain particles according to $N(r) \sim r^2$, and $N(r) \sim r^{0.8}$. (b) Relative frequency of strain values as a function of strain magnitude for particles with distance $r<10 \ \mu m$ (blue) and $r>20 \ \mu m$ from the center (red). The dashed lines indicate fits to a Gaussian (red) and a power-law distribution (blue).

This fractal structure of deformation allows plasticity to penetrate the glass at a low density, and to produce macroscopic flow at low energetic cost. While our limited field of view allows us to elucidate this scaling of strain only for a limited range of length scales, we obtain further evidence of the fractal nature of deformation from the probability distribution of strain. We show histograms of strain values in Fig. 6.8b, where blue and red
dots indicate particles in and outside the central zone, respectively. The data reveals the very different character of the distributions: inside the central zone, particles exhibit a Gaussian strain distribution (blue dashed line), indicating that plasticity occurs in a statistically independent manner. In contrast, particles outside the central zone exhibit a power-law distribution (red dashed line), indicating strong correlation, in agreement with the fractal structure of deformation. We hypothesize that the high shear stress in the centre is sufficiently strong so that plastic zones can form independently of each other, while at much weaker stresses further away from the centre, plastic zones can only form when triggered by adjacent plastic zones, and hence plasticity occurs in a highly correlated fashion. These strong correlations result in a hierarchical structure of deformation at the onset of plasticity.

Recent theoretical work on disordered fiber networks [6] indicates that the loss of network rigidity is associated with diverging strain correlations and a true critical point. Our observation of a hierarchical strain distribution indicates that even a glass shows hallmarks of critical behavior at the onset of plastic flow. Critical points demarcate qualitative changes of behavior, typically associated with symmetry changes. In the incipient deformation investigated here, the symmetry change occurs in time, and demarcates the transition from reversible elastic to irreversible plastic deformation.

6.5.5: Spatial correlation of strain field

To investigate this critical behavior of incipient deformation in more detail we investigate how the strain is correlated in space. To do so, we determine the spatial strain correlation function:

$$C(\Delta r) = \frac{\langle \varepsilon(r) \varepsilon(r + \Delta r) \rangle - \langle \varepsilon(r) \rangle^2}{\langle \varepsilon(r)^2 \rangle - \langle \varepsilon(r) \rangle^2}$$ (6.13)

that correlates values of strain at location separated by $\Delta r$. The result is shown in Fig. 6.9a for different times of indentation. These data seems to indicate a power-law relation of early times when the needle touches the colloidal suspension or very small amount goes inside. With ongoing indentation the power-law relation vanishes and the deformation is
governed by the deformation in the centre. This leads to higher correlation amount, but a faster decay at larger distances.

![Image](image_url)

**Figure 6.9:** (a) Correlation function of strain as a function of particle separation according to eq. 6.13. The dashed line is to guide eyes (b) Correlation between local shear strain and local free volume, as function of time (eq. 6.14).

### 6.5.6: Connection to the glass structure

According to the free volume models proposed by Argon and Spaepen [11, 18], the formation of a local shear strain is associated with significant dilation. To test these ideas, we study the correlation between local strain and free volume. Using the position of particles that are identified from the confocal images, we calculate the free volume for each particle which is assumed to be confined to its Voronoi cell. Following [19] and consistent with [20, 21], the free volume associated with a Voronoi cell is defined as the volume of a smaller cell generated from the Voronoi cell by moving the faces normally inside of a distance $R_p$, where $R_p$ is the particle diameter. It should be noted that the assumption of considering that the particles are confined in cells is valid only at high densities where the particles are densely packed that no rearrangements happen on the
time scale of obtaining the configuration. We compute the correlation between shear strain $\varepsilon$ and free volume $V_f$ using:

$$C = \frac{\sum_N (\varepsilon(r) - <\varepsilon(r)>)(V_f - <V_f>)}{\sqrt{\sum_N (\varepsilon(r) - <\varepsilon(r)>)^2 \times \sum_N (V_f - <V_f>)^2}}$$  \hspace{1cm} (6.14)

We plot $C$ as a function of time in Fig. 6.9b, where we have averaged over all particles $N$ in the slice. The values that we obtain fluctuate around 0.06. This value clearly demonstrates a weak correlation between the shear strain and the free volume.

**6.5.7: Time evaluation of elastic energy**

Using the shear modulus determined in Sec. 6.4, we can compute the local elastic energy of a particle and its nearest neighbour distortion assuming that a linear approximation still holds. By using Eqn. 6.10, we thus calculate the elastic distortion energy associated with the strains $\varepsilon_{ij}$.

![Figure 6.10: Time series showing the evolution of the total strain energy](image)

*The time evolution shows the heterogeneous distribution of the elastic energy. High-energy regions are depicted in dark red; these grow in time and finally join each other.*
We show the resulting distribution of energies and their time evolution in Fig. 6.10. The colours illustrate elastic energy magnitude. The figure shows the strong heterogeneity of the elastic energy associated with the heterogeneous strain field.

6.6: Discussion and conclusion

The application of indentation on a colloidal glass reveals a surprising hierarchical structure of deformation at the onset of permanent deformation. This hierarchical structure is reminiscent of a second-order transition, indicating that the glass at the onset of flow exhibits signatures of criticality, in agreement with recent theoretical work on fiber networks. The similarity between the fractal structure of deformation and that encountered in the aggregation of particles suggests an analogy between the mechanisms of structural arrest and shear-induced relaxation. While the direct imaging of microscopic strain allows us to visualize its fractal structure in a colloidal glass, we expect our results to be general and apply to molecular glasses as well. We estimate from approximate conversion of the deformation rate from indentation to uniaxial strain rate that the strain rate of our experiment is \( \sim 3 \cdot 10^{-5} \text{ s}^{-1} \), a factor of three larger than the inverse structural relaxation time of our glass. This ratio is typically encountered in the high-temperature deformation of molecular glasses [5,18]. For example, for typical strain rates employed in the nanoindentation of atomic glasses in the range of \( 10^{-3} \text{ s}^{-1} \) to \( 1 \text{ s}^{-1} \), the same ratio of imposed strain rate to relaxation rate is found in the range between 0.9 and 1.1 of the glass transition temperature. Deformation tests on typical metallic glasses show that indeed homogeneous deformation occurs at these strain rates and temperatures, in agreement with our experiments [5]. Therefore, our observations should be relevant for these atomic glasses as well. The long-range nature of the strain field should then have important consequences for the deformation in small dimensions, such as the one exploited in nano and micron-scale mechanical devices.

References