Vibrations in materials with granularity

Zeravcic, Z.

Chapter 1

Introduction

Light, the sound of G#, earthquakes, gravitational waves, water waves, microwaves... visible or not, waves are everywhere around us. We are dependent on waves: without them we would not be able to communicate or to perceive our environment. Their ubiquity makes waves and wave phenomena some of the most important and studied constituents of the physical world.

The nature of a *wave* depends on its physical origin, and here our focus is going to be on mechanical ones. Perturbations propagating in space and time within a medium (*i.e.*, *mechanical waves*) usually transfer energy between distant points but, for small enough vibration amplitude, do not involve a lasting displacement of the elementary particles of the medium. The building blocks, however big or small, instead *vibrate* around their rest positions. Coupling of the elementary constituents of the medium enables a vibration of one constituent to induce an oscillation in another one. Therefore vibrations of elementary constituents constitute an emergent wave in the medium.

The literature on waves and vibrations in simple systems and ordered structures is vast. Once we move to disordered media, however, a whole new world of (non-) intuitive phenomena emerges, and becomes a continuing source of exciting and challenging new problems.

In general, the presence of disorder in materials can lead to fundamental changes in mechanical, thermodynamic, static and dynamic properties. In particular, waves in the material can completely change their character, and details of the nature of vibrations become an important window into the physics of such materials. Furthermore, understanding the properties of vibrations is crucial for entire classes of materials which are dominated by disorder effects, but elude a simple description within

the standard methods of solid state and statistical physics. A prominent example are glasses, which still pose numerous long-standing puzzles, as they are systems intrinsically out of equilibrium.

This thesis is a journey throughout the world of *materials with granularity*, by which we mean materials consisting of particles large enough, such that the temperature does not play any role in the dynamics of the system. More specifically, we will study properties of vibrations of systems consisting of collections of *jammed* spherical and ellipsoidal particles, as well as clusters of interacting bubbles. But before we embark, we will spend some time in this Chapter to remind the reader of some of the properties of vibrations in classical solids. We will also give a basic introduction to the concept of jamming and mechanical properties of sphere packings and sound propagation in clusters of bubbles.

1.1 Vibrations in Classical Solids

Classical solids represent a state of matter in which the motion of each constituent element is confined to a small volume around its equilibrium position. The simplest example is a crystal, where the equilibrium positions form a lattice in space. We will keep the picture of a crystal in mind throughout this Subsection, since it is the easiest to describe.

Most physical properties depend on the possible excitations of such a system, because these are probed by any external perturbation during a measurement. When studying mechanical properties, the movement of atoms (or molecules) is the basic degree of freedom that is excited (translational and/or rotational). When displaced from its equilibrium position, an atom feels a restitutive *elastic* force towards its equilibrium position, originating in the interaction with its neighbors, and therefore performs oscillations around it. These vibrations form the elementary excitations of the solid in this context.

1.1.1 Dynamical matrix

The energy of a given state depends on the displacements of the atoms around their equilibrium positions. Throughout this thesis, we will focus on small-amplitude vibrations. Irreversible (plastic) rearrangements are not considered. Since the displacements are small (the solid is "robust"), we can expand the excitation energy in a Taylor series in the displacements [1]. Because the equilibrium state is a minimum of the potential, all the terms linear in displacement vanish. The rest of the terms give the *potential energy* (of elastic displacement), which is the starting point for our analysis.

More explicitly, the expression for the potential energy in the harmonic approximation for the dynamics, *i.e.*, up to quadratic terms in the displacement, is:

$$U = \frac{1}{2} \sum_{\substack{i,j \\ \alpha,\beta}} u_i^\alpha H_{i,j}^{\alpha\beta} u_j^\beta, \tag{1.1}$$

where u_i^α is the generalized displacement of particle at site i, and α in general enumerates the degrees of freedom of the particle. In the simplest case the particle can only translate. We will, however, also give them the freedom to rotate or dilate. The generalized displacements u_i^α will represent displacements or angles or particle radii, while masses m_i (see below) will also denote moments of inertia. The matrix appearing in the previous equation consists of second derivatives of the potential:

$$H_{i,j}^{\alpha\beta} = \frac{\partial^2 V(\vec{u})}{\partial u_i^\alpha \partial u_j^\beta}, \tag{1.2}$$

is the *Stiffness or Hessian Matrix*. In the language of elasticity, this is a matrix of spring constants. This matrix appears in the equations of motion (we give the example of spherical particles with different masses):

$$m_i \ddot{u}_i^\alpha = -\sum_{j,\alpha} H_{i,j}^{\alpha\beta} u_j^\beta, \qquad i = 1,...N. \tag{1.3}$$

We define the *Dynamical Matrix* $\hat{D}$ to be such that the equations of motion can be written in a simple matrix form $\ddot{u} = -\hat{D}u$. The precise form of $\hat{D}$ in this example is:

$$D_{i,j}^{\alpha\beta} = \frac{1}{\sqrt{m_i m_j}} H_{i,j}^{\alpha\beta}{}^1 \tag{1.4}$$

Note that the diagonal elements of the Hessian, *i.e.*, self-interactions, $H_{ii}^{\alpha\beta}$ are undefined. However, we know that translation or rotation of the entire system does not change the energy of the system. Imposing this as a condition for the equations of motion gives the values of the diagonal elements. In the above example (when there are no angular degrees of freedom) the diagonal elements are:

$$D_{i,i}^{\alpha\beta} = -\sum_{j\neq i} D_{i,j}^{\alpha\beta}. \tag{1.5}$$

We will use the Dynamical Matrix intensively throughout this thesis as the starting point in the analysis of our systems. In typical solid state systems the interactions are effectively short-ranged, hence the dynamical matrix is sparse. This will not be true in the bubble vibrational problem of Chapter 4, however, where the interactions originate from the long-ranged pressure field. Note that in problems like glasses, it is highly non-trivial to write down an appropriate Dynamical Matrix because of the disorder in the position of the constituents.

1.1.2 Vibrational spectrum

The fundamental step towards identifying the elementary excitations of a given solid is solving the equations of motion. These take the form $det|\hat{D} - \omega^2| = 0$, where we

[1] Once particles are non-spherical, rescaling factor will contain moments of inertia and factors due to use of non-linear coordinates (see Chapter 2).

assumed that atoms vibrate with frequency ω, *i.e.*, we assumed solutions in the form $u_i^\alpha \sim e^{i\omega t}$. The result of the diagonalization of the Dynamical Matrix is a set of eigenvectors, or eigenmodes $\vec{q}_a = \{q_a^{i,\alpha}\}$, which are linear combinations of displacements of individual atoms within the solid. The potential U becomes a sum of independent harmonic (eigen)oscillator potentials, $U = \frac{1}{2}\sum_{a,i\alpha}\omega_a^2\left(q_a^{i,\alpha}\right)^2$. That means that the a-th eigenvalue of $\hat{D}$, *i.e.*, the coefficient of the potential energy of the a-th eigenmode oscillation, is its frequency squared, ω_a^2.

Eigenmodes are the elementary excitations of the solid we have sought! They are independent and mutually orthogonal, *i.e.*, if we were to excite one of the modes, it would continue to oscillate in an unchanged form indefinitely. (This is true within the harmonic approximation we are using, in which phonon-phonon and electron-phonon scattering processes are neglected). In the present context of the vibrations of solids, these eigenmodes are called *phonons*.

The set of eigenfrequencies $\{\omega_a^2\}$ represents the spectrum of vibrational excitations of the solid. The number of modes is equal to the total number of the degrees of freedom N, *i.e.*, $a = 1...N$, and therefore the spectrum becomes dense in the limit of large systems. We can now ask how many modes there are in a given interval of energy or frequency. This leads to a definition of the normalized *Density of States* $ND(\omega)$. In the interval $(\omega, \omega + \mathrm{d}\omega)$ there are $ND(\omega)\mathrm{d}\omega$ modes. Since many properties of the solid depend on its vibrational modes and the energy, the density of states (DOS) is a central quantity in determining the behavior of the system.

Debye scaling

Let us now consider in more detail the case of a simple monatomic crystal, and the behavior of its low energy $D(\omega)$. We focus on the low energy part of the spectrum, since it has universal properties independent of the details of the crystal lattice. This follows from the translational symmetry breaking, which occurs when the atoms in a liquid state form a solid by occupying discrete positions in a lattice.

Phonons in crystals take the form of plane waves, *i.e.*, they are periodic in space with a fixed direction of propagation given by the wave-vector $\vec{k}$. In $3d$ there are two transverse phonons (the vibration is orthogonal to $\vec{k}$) and one longitudinal phonon (vibration is parallel to $\vec{k}$) for each given $\vec{k}$, since each atom has three degrees of freedom.

Since phonons are the sound waves in the crystal, at small momentum $|\vec{k}|$ their frequency in terms of momentum (*i.e.*, the dispersion relation) is given by $\omega(\vec{k}) = c(\hat{\vec{k}})|\vec{k}|$, where c is the anisotropic speed of sound. To now find the $D(\omega)$, we just use the fact that phonon states are counted by their momentum value. We take the system to be represented by a box with side length L, which constrains the allowed values of momentum to $k_i = (2\pi/L)n$, with $i = x, y, z$, and n an integer. Now the density of states follows from identifying the number of momentum states in a small volume of momentum space, $(2\pi)^3/V\,\mathrm{d}^3\vec{k}$, with the number of phonon states counted by their frequency value, $D(\omega)\mathrm{d}\omega$. Using this together with the dispersion relation

that connects ω to $\vec{k}$, one obtains the well-known *Debye* expression for the density of states of low-frequency phonons:

$$D(\omega) \sim \omega^{d-1}, \tag{1.6}$$

where d is the dimension of the system.
This Debye behavior of $D(\omega)$ is very robust and is observed in a variety of real materials, at least indirectly through the low-temperature behavior of the heat capacity $c_V \sim T^d$. However, significant deviations can occur, as we will see later!

1.1.3 Disorder

Disorder is an unavoidable feature of all materials. There are classes of systems, including some we will consider in this thesis, where disorder is the essential property defining the system. There are also various types of disorder. In Chapter 4 we will meet several of them, first acting in the system simultaneously, and then analyzed separately. In the example of a crystal, we could imagine the spring constants becoming random (influencing interaction of particles), but could also consider just randomizing masses of atoms (changing their inertia).

The central question for us is: What happens to waves in disordered systems? Just how robust is the Debye behavior of the vibrational DOS? The search for answers to these questions has been at the forefront of research for at least 50 years, and here we will take a quick look at the central notions important for this thesis.

First concerning the DOS very generally, let us note that introducing disorder causes the smearing of sharp peak features (van Hove singularities) which are a hallmark of the underlying lattice. In granular materials however, we will consider a state near to a phase transition, which has the additional critical property of being barely mechanically stable. This has an even more dramatic influence, leading to the failure of the Debye model, as a plateau of low frequency modes develops in the DOS. Such behavior is also observed in experiments on glasses.

1.1.4 Localization

If we imagine disorder centers introduced in a homogeneous medium, we can understand that they will cause our waves to scatter multiple times between them. It has been found [2,3] that this will result in a *diffusive* propagation (*e.g.*, of energy), and of course the loss of well defined momentum $\vec{k}$. This was novel behavior at the time of discovery, not expected from a wave equation like the one governing phonons. Even more interesting, the wave can experience *weak localization* when, due to interference effects, it has an increase in backscattering probability, and the diffusion slows down [2].

In 1958, by studying the problem of quantum mechanical electron wave propagation, Anderson realized that diffusion can actually stop completely. This is called *strong localization* of the wave pattern [4]. The result is a pattern localized in space,

with an envelope that falls off exponentially with distance, thereby defining a *localization length* ξ.

It turns out that dimensionality plays a decisive role in localization. Heuristically, going back to the concept of diffusion, we can imagine it as representing a particle that is taking a random walk through our system. The probability of *finding the particle near the origin after a long time* is finite in $1d$ and $2d$, but goes to zero in $3d$ (in the limit of infinite system size), leading to qualitatively different backscattering [2]. The outcome is that in $1d$ and $2d$, all waves are *strongly* ("Anderson") localized in an infinite system with arbitrarily weak disorder. It was shown by John *et al.* [5] that in the case of short range uncorrelated mass disorder the localization length diverges as $\xi \sim 1/\omega^2$ in $1d$ and $\xi \sim e^{1/\omega^2}$ in $2d$ as the translational modes at $\omega = 0$ are approached. In $3d$, however, one can find a localization-delocalization transition at a finite frequency.

Our focus is on vibrational modes in both $2d$ and $3d$, and their experience of these types of effects. An important realization is that systems are usually not infinite, certainly not in computer simulations. This means that the system size might be too small to uncover the localized nature of a state (a vibrational mode) even in $2d$. In other words, it can happen that the mode spans the entire examined system, and maybe even takes a shape resembling a plane-wave, but is actually *intrinsically* localized on a scale which happens to be larger than the particular system size L. In Chapter 3 we will introduce a method able to discern the localization length ξ, even when in the regime of $\xi > L$.

Finally we want to address the issue of interaction strength. In vibrational problems of solids, it is natural to consider only nearest-neighbor interactions (the Dynamical Matrix, *i.e.*, "springs", connect only nearest-neighbors in the lattice); this was also the case in Anderson's considerations, where electrons where able to hop only between neighbor lattice sites. In Chapter 4 however we will consider a system of coupled bubbles with *long-range interactions* that fall off as $1/r$ with distance. We will see that the switch from short- to long-range interactions leads to *power law* localized states, instead of the usual exponential form characteristic of strong localization.

1.2 Vibrational modes in granular matter near Jamming

The author of this thesis cannot resist introducing the intriguing aspects of granular systems in the following illustrative way.

Imagine that you have cravings for pancakes, and you decide to make them. You walk into the kitchen and start gathering the necessary ingredients, among which is flour. What you are not aware of yet is the beautiful physics hidden in that bag. Flour, as a type of granular material, can behave: *(i)* solid-like, since it will support your weight, if you step on it, *(ii)* liquid-like, since it will pour out of the bag into the mixing bowl, and *(iii)* gas-like, since you will start to sneeze or cough because of the fine white powder in the air.

As illustrated in the above paragraph, we encounter granular matter on a daily ba-

sis. Even more importantly, materials in this form are the second most used in industry, right after water [6]. Although the thorough understanding of granular systems therefore has vast relevance for practical applications, they have been the center of much scientific attention in the past decade because they seem to exhibit properties characteristic of glasses. They have proved themselves as a useful working model in the description of some properties of glasses, limited by the fact that granular systems are athermal.

The study of granular matter covers numerous static properties [7–36], as well as rheological phenomena [37–43], explored in a variety of theoretical, numerical and experimental models. Model systems consist of a range of particle types (*e.g.* spheres, asymmetric particles...) immersed in various solutions (*e.g.* air, liquids...) and under diverse external conditions (*e.g.* forcing, flow...). As will be explained in more detail below, one of the central phenomenon exhibited by all granular materials is the transition from liquid- to solid-like behavior with increasing density. The appropriate name of *jamming* should remind us of a traffic jam, the point at which flow becomes arrested.

1.2.1 Jamming idea

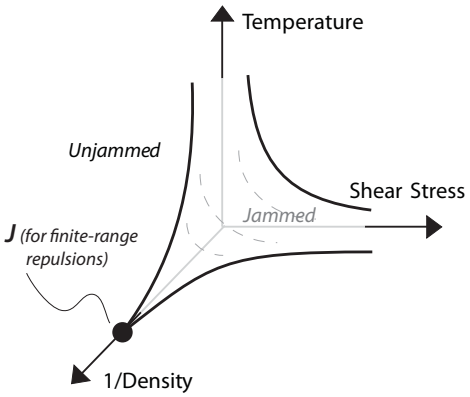

Figure 1.1: A sketch of a jamming phase diagram.

In 1998. Liu and Nagel put forward the idea to view diverse amorphous materials — foams, colloids, emulsions, molecular glasses and other materials with granularity — in the context of the *jamming phase diagram*, Fig. 1.1, [44]. All these materials jam into rigid, disordered states, as different mechanical and thermodynamical variables are changed. Flowing foams can exhibit the *jamming transition, i.e.*, flow will be arrested, if the shear stress is reduced below the yield stress σ_y. Another example of a rigidity transition would be the transformation of supercooled liquids into glasses once the temperature is lowered below the glass transition temperature T_g. On the

other hand, flowing grains will jam as the density is increased above some critical density ϕ_c, usually identified with the random close packing density [7, 11].

A canonical model system for studying properties of these disordered materials are packings (*i.e.*, collections) of soft, repulsive, frictionless spheres that interact only when they are in contact. Throughout this thesis we will focus on packings that reside on the zero temperature and zero shear stress axes of the jamming phase diagram, which represent models for granular materials, foams and non-brownian emulsions. Their transition to rigidity is governed by changes in the packing fraction ϕ.

The beauty of soft frictionless sphere packings is that for a given protocol used to make packings, they exhibit the jamming transition at a well defined density ϕ_c. Another way to characterize this transition is by saying that a packing is jammed once the confining pressure p becomes positive (this is independent of the protocol used, hence a more precise measure of a jammed system). The point in the jamming phase diagram at which $T = 0$, $\sigma = 0$ and $p = 0$ is termed as *point J* (Fig. 1.1). At this point the average contact number Z (*i.e.*, average number of contacts per particle in a packing) reaches the so-called *isostatic value* Z_{iso}. As we will see in more detail in Sections 1.2.3 and 1.2.5, the very rich behavior these systems exhibit is intimately related with the geometry of the underlying contact network, *i.e.*, distance to this isostatic value. As we will see in subsequent Sections, at $Z_{\rm iso}$ these systems are marginally stable, *i.e.*, they are *marginal solids*.

1.2.2 Density of States

As we introduced in Section 1.1.2, one of the most robust behaviors in material science is that of the Debye behavior of the low-frequency range of the vibrational density of states. However, this law breaks down in a fascinating manner for the case of a solid made by jamming spheres [7, 18]!

To analyze the vibrational modes of our granular packings, we need to form and diagonalize the dynamical matrix $\hat{D}$ that we introduced in Section 1.1.1. The eigenfrequencies obtained for packings at different distances from the jamming point are shown in Fig. 1.2. Let us first focus on what looks familiar. The dashed black curve shows data considerably above the jamming point (see Fig. 1.2(1)), where one can see the Debye-like behavior of the low-frequency range of the $D(\omega)$. Once we start decreasing the density ϕ towards point J (see Fig. 1.2(2)), an excess of low-frequency modes starts to appear, suppressing the Debye behavior (solid black line). This plateau extends to a finite value for $\omega \to 0$ at the jamming point, meaning that compared to a "normal" (Debye-like) solid there are far more ways with which one can "move" all the particles in the system around their equilibrium positions with little energy cost! Modes belonging to the plateau in the DOS are usually called "soft modes". The existence of a low-frequency plateau in $D(\omega)$ is a hallmark of marginal solids, *i.e.*, solids that are on the verge of instability.

In glasses one universally observes an intriguingly similar enhancement of low-energy excitations (the "boson peak"). In those materials, a long-standing candidate explanation is the picture of abundant two level units within the material, which can

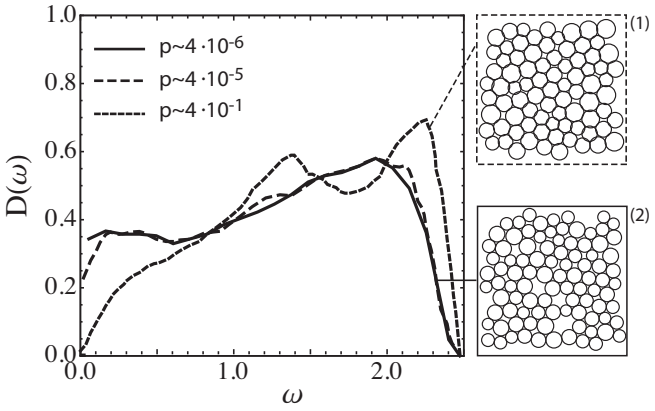

Figure 1.2: The density of states for $2d$ harmonic packings at different distances to the jamming point. Note how the Debye like behavior of highly compressed packing (dashed line) transforms into a plateau of low-frequency excitations on approach to the jamming point (solid black line).

easily "flip" between the two states at low energy cost [45–47]. It is exciting to consider the possibility of understanding the appearance of these two level states in terms of the low energy vibrational states of non-spherical particles, like the ellipsoids we consider in Chapter 2.

1.2.3 Maxwell Rigidity Criterion

We have now mentioned phrases like *marginal solid* and *isostatic packings* several times. This section is therefore devoted to explaining these concepts, which can be traced back to Maxwell. The explanation is based on global counting arguments of the number of degrees freedom needed to constrain all the available motions of the system. As the reader will see, understanding these ideas is crucial for grasping the origin of the excess low-frequency modes and some of the anomalous scalings we will present in the subsequent Section.

According to Maxwell [48, 49], *rigid* means that all the available motions in the system are constrained. Let us start from a jammed packing of N spherical frictionless particles in d dimensions that have on average Z *non-trivial* contacts with their neighbors[2]. The relevant degrees of freedom of these particles are translations in d dimensions, hence we have dN degrees of freedom. Since every contact is shared between two particles, and a particle has Z neighbors on average, the number of

[2] We are emphasizing the word non-trivial here because packings can have particles that are not in force-carrying contact with the rest of the particles in the packing. The colloquial term used for these particles is "rattlers" or "floaters". They should be left out from the above counting arguments.

contact forces is $ZN/2$, which is equal to the number of non-zero terms in the potential energy. The potential energy is a function of all the dN non-trivial degrees of freedom. Since we are jammed (there are no available motions in the system that cost zero energy!), the number of terms in the potential energy exceeds the number of degrees of freedom, *i.e.*, $ZN/2 \geq dN$, which gives $Z \geq 2d$. On the other hand, at the jamming point ($p \downarrow 0$) particles are undeformed, which means that the distance between touching particles has to be equal to the sum of their radii. The "touching" condition then yields $NZ/2$ constraints for dN non-trivial degrees of freedom. In a generic packing, there are solutions when $Z \leq 2d$.

Combining the two obtained inequalities, both valid exactly at the jamming point, we get the average coordination number of an isostatic solid:

$$Z_{\text{iso}} = 2d. \tag{1.7}$$

This result is very robust: it does not vary with the details of the interaction, polydispersity or protocol for making the system. As we shall see nearly all the beautiful behavior observed for granular packings can be related to the distance to the isostatic point, *i.e.*, $\delta Z \equiv Z - Z_{\text{iso}}$.

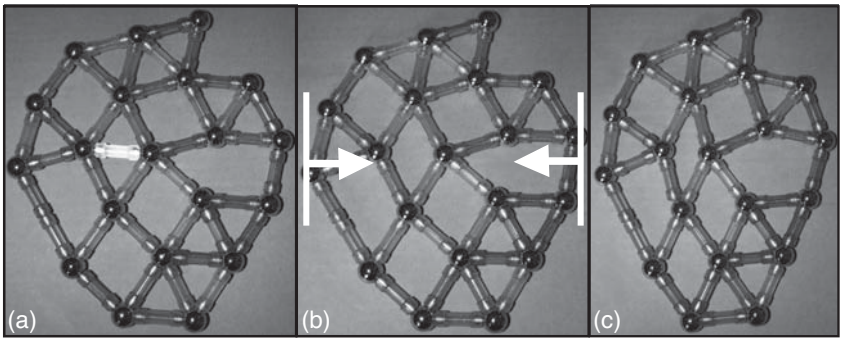

Figure 1.3: (a) A bar-and-joint framework. The system is rigid: global translations and rotation are the only available motions of the joints that do not stretch or compress any of the bars. If we remove one bar (marked), rigidity is lost. (b) By pushing on the edges of the framework (arrows), the system can be deformed non-trivially at zero energy cost, *i.e.*, none of the bars change length. (c) The new state after deformation. This example is directly relevant to disk and sphere packings because every such packing can be mapped directly to a bar-and-joint framework.

The above result tells us that a packing at the isostaticity point has just enough contacts to be stable (hence the expression *marginally stable*). Note that this is a *global* condition, and it differs from the *local* stability condition, according to which each particle needs $d+1$ contacts to have a fixed position. A natural question is then what happens if we have fewer contacts than needed to maintain rigidity. A simple

sketch of the answer to this question is shown in Fig. 1.3. Imagine that our packing has one contact less than required. By the argument above, we expect that this permits one rearrangement of the particles that will not change any distances between them. By removing the contact, we have created a *zero energy* mode (in literature sometimes called a "floppy" mode) in the system. Note that these are global modes, since all the particles will be involved in the rearrangement if their relative distances are to stay fixed! Later, floppy modes will help us identify the actual low-frequency soft modes that populate the DOS plateau (see Section 1.2.5).

The Maxwell argument can be extended to the case of particles with additional degrees of freedom, for example non-spherical particles that can also rotate around their center. We will explore the case of axisymmetric ellipsoids, and ellipses, in Chapter 2. There we will need to revisit the meaning of the Maxwell criterion, and settle the question of how $Z_{\rm iso}$, and the accompanying vibrational density of states, change as we gradually distort spheres to ellipsoids and introduce non-trivial rotational degrees of freedom. We will find that the Maxwell criterion cannot capture the continuous change of $Z_{\rm iso}$ with ellipticity, but the introduction of a rotational band into the DOS does leaves the main features of the jamming point scenario intact.

1.2.4 Packings

Over the past decade, many mechanical and dynamical properties of granular materials have been studied as a function of the distance to the jamming point. As we already mentioned, the model system that is used to probe these properties are packings of repulsive frictionless soft spheres or discs, that interact only when they are in contact. The force that two particles in contact exert on each other is a function of the overlap between them, which is defined as $\delta_{ij} = R_i + R_j - r_{ij}$. R_i and R_j are the radii of the ith and jth particle respectively, and r_{ij} is the distance between their centers. Force models one commonly encounters are: *(i) one-sided harmonic springs*, for which the repulsive force is linearly proportional to the overlap, $f_{ij} \sim \delta_{ij}$ and *(ii) Hertzian springs*, for which $f_{ij} \sim \delta_{ij}^{3/2}$.[3]

Once the force law is chosen, the aforementioned packings are made using protocols like *molecular dynamics* [10, 14, 50, 51] and *conjugate gradient method* [7]. In the former, the simulation starts from a loose gas of particles which are gradually compressed (either by shrinking the container, or increasing their radii) until they reach a jammed state. The system cools down through inelastic collisions or viscous drag. The conjugate gradient (CG) method is based on the fact that a stable packing is in a minimum of elastic energy (1.1). Starting from a random configuration (*i.e.*, Poisson point process, or $T = 0$ state), the system is brought to the nearest potential energy minimum. In this thesis (Chapters 2 and 3) we used packings made with both protocols.

In order to give the reader an idea of the kind of systems we are studying and

[3] For the case of Hertzian force law, contacts effectively become stiffer as the particles becomes more compressed.

how states far and close to the jamming point differ, Fig. 2.2 depicts packings, force networks, density of states and response to shear at two different pressures p.

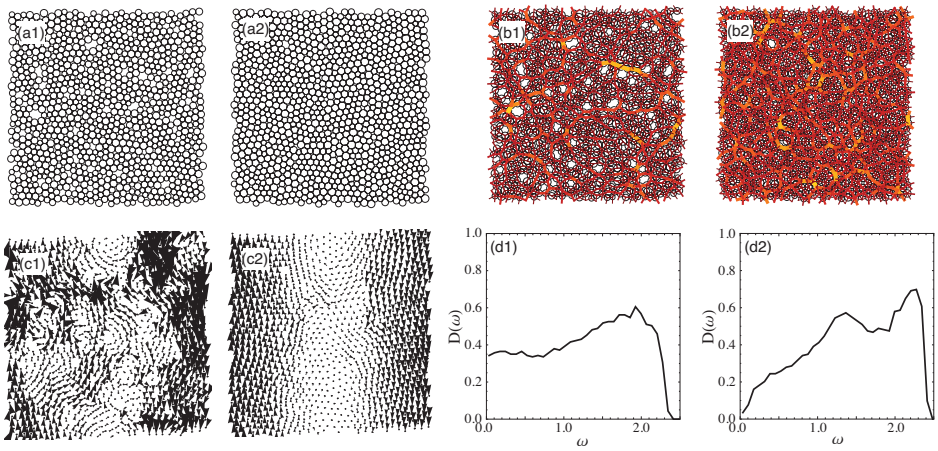

Figure 1.4: Illustrative example of different properties of granular packings for two different pressures: at the jamming onset (a1-d1) and in the classical solid regime (a2-d2). (a1) and (a2) are visualized packings, with rattlers removed. (b1) and (b2) show the force networks. Force color different than red marks bonds with large forces. (c1) and (c2) show the response to shearing of the two extremal packings. Note the non-affine pattern for the case of just jammed packing. (d1) and (d2) are the corresponding density of states. Close to the jamming point a plateau of low-frequency modes develops (see main text for details), whereas far from jamming, low-frequency part of $D(\omega)$ resembles the Debye behavior.

1.2.5 Anomalous scalings

In this Subsection we will give a flavor of the critical nature of the jamming point, by looking at the response of granular packings to shear, and the scaling of the average contact number Z (*i.e.*, average number of contacts per particle within a packing). The Z is the *order parameter* of the jamming transition: it is zero on the unjammed side[4], and has finite values on the jammed side. For a detailed introduction to many other interesting behaviors, see the recent review by van Hecke [52].

The behavior of the average contact number Z near the jamming transition (Fig. 1.5(a)) defies intuition. Let us start from a packing close to the jamming point and compress it by 1% (*i.e.*, we change the packing fraction ϕ by 1%). Assuming affine

[4] In a dilute system particles can be in contact, but to minimize the energy these contacts are not force-carrying. Technically, all particles are rattlers.

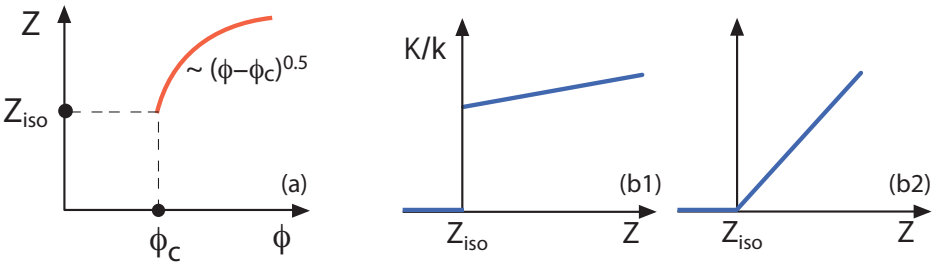

Figure 1.5: Anomalous scalings. (a) Sketch of the change of the average contact number Z with the change of density ϕ. This order parameter shows a discontinuous jump, reminiscent of the first order phase transition. (b1-b2) Scaling of the elastic moduli with the increase of average Z. (b1) shows the bulk modulus, and (b2) the shear modulus. In both (b1) and (b2) k represents spring constant.

deformations[5], one would naively expect a linear increase of δZ — all the particles that were separated by 1% of their radii will now come into contact. However, many studies have shown that the contact number has a universal *square root* dependence on the excess packing fraction $\delta\phi = \phi - \phi_c$, independent of the details of interactions, polydispersity of particles and the dimension of the system! So we have a discontinuous jump in the average contact number Z at point J, and then anomalous scaling as we move away from it! Without going into details let us just state that over the years explanations of this unusual scaling have been proposed, but in the opinion of this author full understanding still does not exist.

The non-trivial power law scaling of the fundamental geometrical property Z is the cause for other anomalous scalings of mechanical properties of the system near jamming. Fig. 1.5(b1-b2) summarizes schematically the universal behavior as it is found in numerical simulations: the bulk modulus K in the units of the contact stiffness k stays constant as δZ is changed, *i.e.*, remains on the order of the spring constant k through the jamming transition; the rescaled shear modulus G/k vanishes linearly with δZ, *i.e.*, inherits the square root scaling with $\delta\phi$, which means the packings become softer in response to shearing.

The same reasoning we demonstrated in the case of the change of δZ with the change of density $\delta\phi$ holds for the elastic moduli — if we compress or shear the system a bit, we are probing the elasticity of the "spring contacts" between our particles, which means that the elastic moduli should not change with $\delta\phi$. But as we saw above, this is true only for the bulk modulus, making the anomalous scaling of the shear modulus seemingly exceptional. Without going into detail, we note that this viewpoint has recently been reconsidered in an attempt to identify what is special about the jammed packings in terms of the scaling of the elastic moduli; namely,

[5] Coordinates change smoothly, following the global deformation.

a reference system to compare to should be random networks. Both G and K for these networks scale linearly with δZ, which is also true only for the shear modulus of jammed packings. The fact that the bulk modulus remains of the order of spring constant at jamming is then to be considered exceptional, and is related to the fact that local displacements *do not* exhibit pure scalings (see [53] for details).

Throughout this Section we have attempted to give the reader some insight into the character of marginal solids. In the end let us go back to the vibrational spectrum of jammed packings, in order to characterize the nature of the low-frequency modes in the plateau of the DOS. The fact that the plateau has a finite value as $\omega \to 0$ implies the existence of a diverging length scale [18].

Isostatic length ℓ^*

Understanding of the excess low-energy modes, *i.e.*, the appearance of a plateau in the DOS of granular systems upon approaching jamming, is obviously of fundamental importance. In this Subsection, we will present the arguments of Wyart [28, 54, 55] about the nature of these low-energy vibrational modes and their contribution to the DOS.

These arguments are based on the *variational* principle. Let us start at the isostatic point. The first step is to construct trial modes which are based on the motions of the system just below the jamming point, and which consequently have zero energies. After "relaxation" these modes become the actual vibrational modes of our system, retaining on their properties. By counting these modes, we will see that the DOS is bounded from below at low ω, *i.e.*, must have a plateau.

The construction starts with cutting (removing) bonds along a boundary, which isolates a subsystem of linear size ℓ in our isostatic system. Because the system was exactly at the isostatic point, each cut contact produces one zero (or so-called "floppy") mode in the subsystem. There are of order ℓ^{d-1} bonds on this boundary, which we have cut, and the same number of zero modes. To build the vibrational modes, we multiply each floppy mode by a sinusoidal envelope that vanishes at the boundary containing the cut bonds. This way we have removed the large motions that occur for particles that have lost a contact. Within the harmonic approximation, the mode has a frequency of order $\omega_\ell \sim 1/\ell$. This is essentially the frequency of the sinusoidal envelope which spans the subsystem, because the underlying floppy mode has energy (frequency) zero.

Finally, assuming that the relaxed, actual, modes in the subsystem will inherit the properties of the Ansatz modes, we can summarize that there are $\sim \ell^{d-1}$ modes of frequency $\sim 1/\ell$ in our subsystem of volume ℓ^d. (These low-energy modes are also called "anomalous", since they appear near isostaticity, and have no resemblance with plane waves.) By using the definition of the DOS, $\int_0^{\omega_\ell} d\omega D(\omega) \sim N_\ell / V_\ell \sim 1/\ell$, we see that $D(\omega) \sim const$ (if $D(\omega)$ can be written as a power law for small ω). There is a plateau in $D(\omega)$ independent of the dimension!

In Fig. 1.6 we demonstrate the above ideas at work. Panel (b) shows the "boundary" along which we remove the particle contacts. The resulting floppy mode is

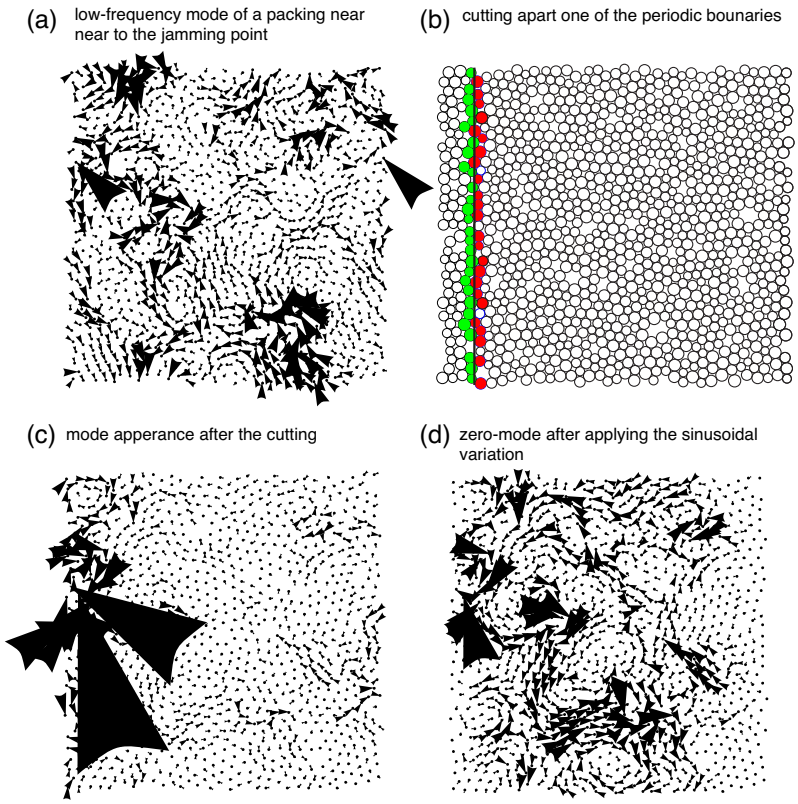

Figure 1.6: Sketch of Wyart's argument: (a) An example of a low-frequency mode. (b) Marked boundary along which crossing contacts will be removed. (c) Resulting floppy mode after removing n contacts. (d) Resulting floppy mode, after applying the sinusoidal modulation.

shown in panel (c), where one can identify the unphysical large displacements that occur on the boundary with severed bonds. In panel (d) however, the floppy mode is multiplied by a sinusoidal envelope that vanishes at the cut, and we can now discern the features of the mode away from the boundary. This ("Ansatz") displacement field is very disordered, controlled by the isostaticity, and should resemble the actual anomalous low-frequency modes that occur in the plateau of DOS of our system. The reader can be convinced of this by looking at panel (a) which shows an actual low-energy vibrational mode, having a shape very similar to the Ansatz in panel (d).

The questions that now arise are what happens at a finite pressure (away from isostaticity). One would expect that at sufficiently large length scales, the response of the packing is elastic — what sets the scale for the crossover to elasticity? And, is there

a way to characterize this crossover of the plateau (isostatic regime) into the standard DOS (which we now associate with "classical" elasticity). We now sketch the answer to these questions, which involves a crossover frequency ω^* occurring in the DOS, and a corresponding crossover length scale ℓ^* characterizing the anomalous modes.

Let us now use all the preceding ideas in a system with a fixed excess contact number per particle equal to δZ. In this case, the total number of excess bonds in the considered subsystem scales as $\delta Z \ell^d$. On the other hand, we create $N_\ell \sim \ell^{d-1}$ degrees of freedom by cutting that many contacts at the subsystem boundary. From the Maxwell criterion in previous subsection, we see that if N_ℓ is less than the umber of excess bonds, we cannot create any floppy modes, which were the source of our trial modes. So, a crossover to the appearance of anomalous modes is given when the two terms (freedom and constraint) are balanced:

$$N_{\ell^*} \sim \delta Z \ell^{*d} \Longrightarrow \ell^* \sim 1/\delta Z.$$

The frequency of anomalous modes at the crossover, *i.e.*, the crossover frequency at which the plateau forms, is accordingly given by $\omega^* \sim 1/\ell^* \sim \delta Z$.

The crossover frequency has been extracted from the DOS of various granular model systems, and is observed to obey the $\omega^* \sim \delta Z$ scaling. This has become one of the hallmark results for granular systems near jamming, and we will establish its meaning and validity for the more complicated case of non-spherical particles in Chapter 2.

From visual inspection of the modes (as in Fig. 1.6(a)), it is apparent that on approach to point J they begin to organize in "swirling" patterns. It is tempting to attempt to extract a length scale from these patterns in the expectation that it will be related or identical to ℓ^*. A fruitful implementation of such an analysis, however, remains elusive [6]. A more productive approach has proven to be that of Ellenbroek and co-workers, who managed to extract ℓ^* directly from the response to mechanical forcing [27].

1.3 Vibrational modes and response to driving in bubble clusters

Finally, we turn our attention to systems of gas bubbles forming a bubble cloud in a liquid. These bubble systems are ubiquitous in nature and application [57, 58]: they appear in underwater acoustics, where underwater noise damping and absorption can occur with the help of bubble clouds or curtains [59, 60]; in the form of bubble curtains they can cause a (repelling) effect on fish schools [61]; they are important for the sound production of colonies of snapping shrimp [62], for submarine detection, for sound propagation in bubbly liquids [63, 64]. In medicine they find crucial application in ultrasound diagnostics with ultrasound contrast agents [65–69], in shock-

[6] Similar patterns are observed in elastic or viscous response to shear. There it appears that the only relevant macroscopic length scale is the system size [43, 56]

wave lithotripsy [70, 71], in ultrasonic and megasonic cleaning [72–74], and also in the process industry.

The bubbles in all of the systems mentioned above can be considered elementary constituents in a material. A single bubble in a liquid, we can understand that it will change size and shape under the influence of the surrounding pressure field. The dynamics of an isolated bubble in such an acoustic field is well understood both theoretically and experimentally (see Chapter 4 and references therein). Considering now a system of many bubbles, the sound emission of an oscillating bubble is felt by the neighboring bubbles: they can attract or repel each other (depending on their mutual size and the external driving pressure [75]).

At this level of detail, we can see the that the bubble clusters fit the paradigm of a classical disordered condensed matter system we introduced in this Chapter. However, there are crucial differences between the jammed packings we have been discussing up to now, and oscillating bubbles, both in the model (equations of motion) and in the physical quantities of interest.

Long range interactions and damping

When we study the vibrational properties of bubble clusters, the distinguishing characteristic of the dynamical equations is that the interaction between the bubbles is *long-ranged*, falling-off as $1/r$ with distance r between the bubbles. This means that each bubble is essentially coupled to all the other bubbles in the system. This is in contrast to the model of grains introduced in Section 1.2, where only neighbors in contact interact!

In condensed matter it is well know that this kind of qualitative change in the interaction can lead to the significant changes in the basic properties of the system (in the example of interacting charged particles, long-range Coulomb force leads to crystallization, *e.g.* colloidal crystals, Wigner crystals, etc.). In terms of the localization behavior of vibrational modes due to disorder, which we have discussed in this Chapter, the long range coupling in bubble clusters makes a qualitative difference, as we will show in Chapter 4.

The second peculiarity of the dynamics of bubbles is the presence of *damping*. Physically, the bubbles dissipate energy as they oscillate because of the acoustic waves they create. Note that loss of energy and damping of motion in a system is common when friction or viscosity are present. However, adding friction to particles in granular packings matters only during the process of making the packing, and does not lead to non-conservative forces in the vibrational dynamics within linear response [76].

The equations of motion, (1.3), acquire an additional term in the presence of damping (in the simplest example of uniform damping):

$$\ddot{u}_i^\alpha = -\sum_{j,\alpha} D_{i,j}^{\alpha\beta} u_j^\beta - \mu \dot{u}_i^\alpha, \qquad i = 1,...N. \tag{1.8}$$

In this simple example the equations again take the form of an eigenvalue problem

of a Dynamical Matrix. However, due to the first order time derivative, the vibrational eigenfrequencies are complex functions of the eigenvalues of the matrix $\hat{D}$. Their imaginary part describes the exponential attenuation of oscillations in time. When describing bubble clusters in Chapter 4, the dynamical equations are more complicated and their solution is more technically involved, so we only demonstrate the principle here.

Response to driving

In the studies of static granular systems near jamming, the primary interest is in understanding the static response of the system, *i.e.*, the elastic moduli, through the description of vibrational modes (not presented in this thesis). In contrast, because of the nature of bubble clusters and the available experimental techniques, one is here primarily interested in the real-time response to external driving. Typically in an experiment there is an acoustic wave applied (representing a periodic driving force), and one tries to understand the time dependent response of the bubbles, which can also be characterized by quantities like the energy absorption or transmission. We will achieve this by analyzing the vibrational properties with the use of condensed matter methods presented in this Chapter. The direct connection between the vibrational states and the dynamical response in the bubble clusters is described in detail in Chapter 4.

1.4 This Thesis

This thesis concerns the vibrational properties of different classical disordered condensed matter systems.

In Chapter 2 we start with vibrational modes of three-dimensional jammed packings of soft ellipsoids of revolution as a function of particle aspect ratio ϵ and volume fraction ϕ. We will find that at the jamming transition for ellipsoids, as distinct from the idealized case of spheres where $\epsilon = 1$, there are many unconstrained and nontrivial rotational degrees of freedom. These constitute a set of zero-frequency modes that are gradually mobilized into a new rotational band as $|\epsilon - 1|$ increases. Quite surprisingly, this new band is separated from zero frequency by a gap and lies below the onset frequency for translational vibrations, ω^*. The presence of these new degrees of freedom leaves unaltered the basic scenario that the translational spectrum is determined only by the average contact number, as introduced in Section 1.2. Indeed, ω^* depends solely on coordination, as it does for compressed packings of spheres. We also discuss the localization properties of vibrational modes in this new band and the regime of large $|\epsilon - 1|$, where the two bands merge.

Chapter 3 is dedicated to the localization of vibrational modes of frictionless granular media. There we introduce a new method, motivated by earlier work on non-Hermitian quantum problems, which works well both in the localized regime where the localization length ξ is much less than the linear size L and in the regime ξ greater

than or of order L, when modes are extended throughout our finite system. Our very lowest frequency modes show "quasi-localized" resonances away from the jamming point; the spatial extent of these regions increases as the jamming point is approached, as expected theoretically. Throughout the remaining frequency range, our data show no signature of the nearness of the jamming point and collapse well when properly rescaled with the system size. Using Random Matrix Theory we derive the scaling relation $\xi \sim L^{d/2}$ for the regime $\xi \gg L$ in d dimensions. To explore our method more, we also calculate ξ for $1d$ disordered chains, $2d$ disordered hexagonal lattice and $2d$ percolation clusters.

The last Chapter addresses the collective oscillations of a bubble cloud in an acoustic field, using concepts and techniques of condensed matter physics. More specifically, we will calculate the eigenmodes and their excitability, eigenfrequency, densities of state, response, absorption, and participation ratio to better understand the collective dynamics of coupled bubbles and to address the question of possible localization of acoustic energy in the bubble cloud. We explore the effects of viscous damping, distance between bubbles, polydispersity, geometric disorder, size of the bubbles, and size of the cloud. For large enough clusters, the collective response is often very different from that of a typical mode, as the frequency response of each mode is sufficiently wide that many modes are excited when the cloud is driven by ultrasound. The reason is the strong effect of viscosity on the collective mode response, which is surprising, as viscous damping effects are small for single bubble oscillations in water. Localization of acoustic energy is only found in the case of substantial bubble size polydispersity or geometric disorder. The lack of localization for weak disorder is traced back to the long-range $1/r$ interaction potential between the individual bubbles. These results are connected to recent experimental observations of collective bubble oscillations in a two-dimensional bubble cloud, where pronounced edge states and a pronounced low frequency response had been observed, both consistent with the present theoretical findings. Finally, an outlook on future possible experiments is given.