Chapter 1

Introduction

1.1. Motivation and significance

Granular materials are collections of discrete, solid particles in ordered or disordered configurations, and are present in many natural and man-made systems. Understanding the fundamental dynamical principles that govern the propagation of stress waves in micro-scale granular systems has implications in many fields of physics and engineering: for example, micro-granular dynamics encompass problems of relevance in powder mixing, acoustics, mining, semiconductor manufacturing, and the pharmaceutical and food industries [1].

Along the broad spectrum of different micro-granular systems, we are especially interested in the characterization of ordered micro-granular systems, and most particularly in micro-granular crystals (which are discrete, ordered arrays of solid micro-particles that are arranged in different lattice geometries). Macroscopic granular crystals, which have been the subject of active research [2-4], have been proposed for use in many engineering applications, such as shock mitigation [5-7], acoustic rectification [8], sound scrambling [9], actuators [10], and acoustic lenses [11, 12]. However, the macro-scale size of the particles tested and modeled in initial studies imposes important limitations to these studies' direct applicability. For example, Donahue et al. [12] demonstrated experimentally the possibility to use properly engineered granular crystals to produce focused, compact pressure pulses in water. Their work directly suggested the use of granular crystals for biomedical imaging and underwater sensing and mapping applications. In these systems, however, the spatial resolution of the propagating pressure pulses, and consequently the size of the focal area, is determined by the size of the particles that compose the granular system. In the experimental setup described by Donahue et al. [9], the particles tested were in the centimeter-scale. This macro-scale dimension limits the spatial resolution of the acoustic pulses traveling through the granular crystals to a few centimeters (and thus necessarily limits the size of the adjacent focal areas to similar dimensions). For acoustic medical imaging or non-destructive evaluation applications, the spatial wavelengths of interest are in the order of a few micrometers. To target these applications it is necessary to miniaturize the granular crystals and to scale the particle size to the micrometer range.

In order to miniaturize granular crystals and explore their functionality at the micro-scale, it is necessary to first understand their underlying physics: Are the assumptions made to model the dynamic response of micro-particles correct? Is the elastic contact interaction still the dominant effect in the contact collisions between two micro-scale particles? How do waves propagate through such miniaturized systems? What are the effective roles of defects/disorder/surface properties and environmental conditions in the dynamics of micro-particles? What role do the particle-substrate interactions play? How do micro-scale granular crystals respond in water? These are some of the fundamental questions we will address in this thesis. Many of these findings extend beyond the limited realm of ordered granular lattices and provide general and fundamental insights into the physical response of micro-scale granular systems.

Despite the fundamental importance of understanding the physics of micro-granular systems, very little experimental work has been conducted at these scales. The lack of experimental investigation results from two major difficulties: first, the absence of reliable methods to assemble micro-particles in controlled configurations (as well as of methods to characterize their precise positions); second, the lack of a systematical way to measure the interaction between micro-granular particles and of the stress propagation through large particle arrays.

Granular crystals are highly nonlinear and discrete. These characteristics are reflected in enhanced practical difficulties in fabricating and mechanically exciting ordered lattices of small-size particles. Conventional experimental techniques, which are widely used in the study of macro-scale granular systems, cannot be directly employed or scaled down to test systems of micro-granules. To provide fundamental insights into the dynamics of micro-particles, it was therefore first necessary to develop a new experimental platform that allowed for repeatable fabrication methods, as well as for mechanical excitation and measurement of micro-granular systems.

We focused on the study of two micro-granular systems: (i) dry, one-dimensional granular systems that consisted of stainless steel micro-particles with a radius of 150 µm, and (ii) wet, two-dimensional granular systems that consisted of SiO_2 particles with a radius of 3.69 µm. In the dry micro-granular systems, we characterized the role of the substrate, the presence of friction, and the mechanics of collisions between two particles. We excited and measured propagation of nonlinear waves along one-dimensional micro-granular chains and studied the influence of inter-particle gaps on the system's group velocity. We further applied this experimental framework to study wave propagation within a self-assembled colloidal system of SiO_2 particles, using both numerical and experimental approaches. We excited the wave propagation within the two-dimensional hexagonal lattice and characterized the role of hydrodynamic interactions within this system.

1.2. Background concerning granular materials

Because of its inhomogeneity, nonlinearity, disorder, and anisotropy, granular material is one of the most challenging subjects in solid mechanics [13, 14]. The experimental difficulties originate from the discrete nature of granular material; the irregularity of sizes, shapes, and materials compositions; and highly nonlinear inter-particle contact forces. The discreteness and the varying particle dimensions together impose difficulty in describing, characterizing, and reproducing granular system configurations. The high nonlinearity in these systems [15] further increases the experimental precision requirement, in which imperfection of the granular system does not even out as the system size grows and sometimes dominates the dynamics response of the system [16].

Due to the experimental difficulties, there are two major trends in the research of granular mechanics. One focuses on the collective and statistical behavior of unstructured granular systems [17-20] without seeking to reproduce exactly the same configuration of samples; it includes the study of granular gas [21-23], granular flow [24-28], particle segregation [29-31], avalanche [32-34], and compression and force in a granular medium [35]. The other approach is to focus on the subsets of granular material, in which the variation in size, shape, and material are limited and particle packing is simplified. One of these subsets is the granular crystal, which refers to highly ordered granular systems.

1.2.1. Ordered granular systems

Research on granular crystals has attracted great attention since the pioneering work done by Nesterenko [2]. He predicted the existence of highly nonlinear soliton-like waves in uncompressed one-dimensional homogeneous granular crystals, specifically an array of spherical particles, where neighboring spheres interact through the Hertzian force. Within the elastic limit of this system, if two neighboring spherical particles have center coordinate x_m, radius R_m, elastic modulus E_m, and poisson ratio ν_m, then the contact force between two particles locating at x_m and x_n is

$$f_{mn}(x_m, x_n) = \frac{4}{3}\frac{\mathrm{E}_m\mathrm{E}_n}{\mathrm{E}_m(1-\nu_n{}^2)+\mathrm{E}_n(1-\nu_m{}^2)}\sqrt{\frac{2\mathrm{R}_m\mathrm{R}_n}{\mathrm{R}_m+\mathrm{R}_n}}\,(R_m + R_n - |x_m - x_n|)_+^{\frac{3}{2}}, \qquad (1.1)$$

where $(R_m + R_n - |x_m - x_n|)_+ = \max(R_m + R_n - |x_m - x_n|, 0)$ is the positive overlap distance of the spheres (if they are not deformed). The equations of motion of the granular chain system are

$$m\,\ddot{x}_n = -\frac{2}{3}\frac{\mathrm{E}}{1-\mathrm{v}^2}\sqrt{\mathrm{R}}\left((2\mathrm{R} - x_{n+1} - x_n)_+^{\frac{3}{2}} - (2\mathrm{R} - x_n + x_{n-1})_+^{\frac{3}{2}}\right). \tag{1.2}$$

Nesterenko solved these equations with long wavelength approximation [2, 3], in which the discrete coordinates of the nth particles, x_n is now redefined as the value of a continuous function of displacement x at position $2Rn$ in a continuous medium, $x_n \equiv x(2Rn)$. He obtained a solution,

$$v(z,t) = \frac{25}{16}\frac{{v_g}^5}{c^4}\cos^4\left(\frac{1}{\sqrt{10}}\frac{z - v_g t}{R}\right), \tag{1.3}$$

where $v(z,t)$ is the particle velocity at $z = 2Rn$, v_g is group velocity, and $c = \sqrt{8ER^3/3(1-\mathrm{v}^2)m}$ is the wave velocity in the material. The results show a highly nonlinear dependency between the group velocity and the maximal amplitude of the particle velocity (maximum velocity),

$$v_g = \sqrt[5]{\frac{16}{25}}\,c^{4/5}{v_{max}}^{1/5}. \tag{1.4}$$

Once the existence of the predicted compact solitary wave was confirmed experimentally [36], numerous numerical [37-39] and experimental [9, 40-43] studies of this system were carried out.

The excellent properties originate from the highly nonlinear interaction, $f \propto \delta^{3/2}$, and it was proven in later years in rigorous mathematics that solitary waves exist in granular chains with arbitrary power-law ($f \propto \delta^n$) nearest-neighbor contact interaction [44-46]. This discovery indicates that the solitary wave is a universal phenomenon in granular medium and that it should exist in a large variety of systems that are made of granular particles with different shapes and types of contact force. It also means that the study of solitary waves in granular materials has fundamental importance in understanding the mechanics of granular materials.

Propagation of soliton waves in one-dimensional granular crystal has since been observed experimentally on several different granular particles, including ellipsoidal particles [47, 48], cylindrical particles [49], hollow particles [50], and heterogeneous media [51-53]. The roles of dissipation [54], plasticity [9, 55] of the granular particles, and velocity tunability under pre-compression [42] have been characterized.

Research interests have advanced to understanding the interaction between two solitary waves [56-60] and between a solitary wave and the interface, boundaries [61], and defects [62, 63] of the granular material. The transmission of solitary waves through the interface between two granular crystals [64] and interface between a granular crystal and elastic medium [10] has also been studied. Yang et al. demonstrated that by sending solitary waves through the interfacing between a known granular chain and an elastic medium, site-specific material properties of the elastic medium can be obtained by measuring the time-delay and reflectivity of the solitary wave [65].

Significant attention has also given to the study of vibration modes and the band structure of granular chains [66]. Despite the highly nonlinear nature of the contact force, a sinusoidal driving can be considered as weakly nonlinear if the background pre-compression force along the chain is big in comparison to the amplitude of the driving force. Experimental and numerical investigations of highly compressed granular chain reveal the existence of band gaps and indicate the possibility of tuning the band gap by tuning pre-compression [67-71]. A particularly interesting case happens when defects (i.e., granular particles with different sizes, masses, or elastic moduli) are placed inside the chain. Boechler et al. conducted numerical and experimental research to reveal the existence of intrinsic localized modes, also known as breathers, in granular chains with defects [72, 73]. An intrinsic localized mode is a localized vibration centered at the defect and amplitude decay exponentially along the lattice. The localized mode has been shown to be universal phenomena in granular chains [16] and can be used for both acoustic switching and rectification [74].

The dynamics of two- and three-dimensional ordered granular systems are relatively poorly understood. While it is suggested that a squared lattice granular system should behave similarly to a one-dimensional granular chain when the solitary wave is propagating along the lattice vectors, a direct generalization of a solitary wave solution to two- and three-dimensional systems has yet been derived. As an intermediate step between one- and two-dimensional granular crystals, Daraio et al. investigated pulse branching and recombination in a y-shaped granular network [75], using the quasi-particle description of a solitary wave to derive transmission coefficients for y-shaped pulse splitting [76, 77]. Leonard et al. further developed an energy mitigation granular network that consists of a three-dimensional network of granular chains [78].

In the early experimental efforts on real two-dimensional granular systems, Shukla et al. used photoelasticity techniques to image wave propagation in various two-dimensional granular crystals, including cubic and hexagonal packing [47, 79-84]. Their experiments show that within higher dimensional granular crystals, the force load path is influenced by the contact angle between lattice elements and wave propagating is altered by the vector connecting the centers of mass of the neighboring particles. The new dimensionality not only introduces more interaction between particles, but also brings new degrees of freedom for designing and engineering the lattice to achieve the desired wave propagating properties. Leonard et al., who studied the wave propagation in two-dimensional square lattices of spherical particles [85], showed that inserting cylindered intruders into these lattices makes it possible to alternate both the wave direction and the energy flux [86].

Despite the good agreement between the average experimental results and wave propagation simulations, the results of experiments involving these two-dimensional granular lattices generally show low repeatability. The difficulty stems from the inability to construct “perfect crystals” in a repeatable fashion. Failing to reproduce perfect crystals has various causes, including the differences of granular particles in size, surface roughness, and shape. The small differences in size and shape do not only change the magnitude of contact force in neighboring particles [87], but they also cause the deformation of the lattice structure and create local compressive areas and gaps [88-90]. Small misalignments that are created by these defects divert force to neighboring particles and therefore scatter the wave propagation [89]. Another mechanism of disorder is through the presence of friction, in which tangential force diverts the wave propagation when the vector connecting the centers of the contacting particles is not parallel to their relative motion [91-93].

Numerical and experimental efforts are devoted to the randomness of granular chains and the influence of this randomness on wave propagation. Manjunath et al. studied random granular chains in which randomness results in the divergence of the magnitude of contact force in relation to neighboring granular particles [94]. They found that the peak amplitudes of propagating waves in random granular chains decay to the degree of randomness with an exponential law, and that the dependency later becomes a power law as randomness further increases. Ponson et al. studied experimentally the effect of randomness on the array of particles. Particles of two different material properties were selected to construct random arrays of diatomic granular crystals. The ratio between two particles defines the randomness of the system, and the researchers observed behavior that was similar to that of the exponential to power law transition [90].

To understand the influence of the polydispersity in higher dimensional systems, load transfer paths are studied numerically in compressed granular crystals [95-98] with different particle sizes. Larger imperfections such as point defects [99, 100] and size deviations [101, 102] of the granular systems have been found to alter the wave propagation more significantly than granular crystals.

1.2.2.Micro-scale granular systems

For particles with diameters of only a few micrometers, the Van der Waals interaction between the particles becomes relatively important. The most famous models describing the influence due to Van der Waals interaction are the Johnson-Kendall-Roberts (JKR) and the Derjaguin-Muller-Toporov (DMT) models [103-105]. In these models, the Hertzian elastic contact potentials are modified to include the electric dipole-dipole energy between particles. The inclusion of these adhesive forces changes the mechanical response of granular systems not only by changing the inter-particle interaction, but also by changing the interaction of the granules with the structures that support the granular assembly [106].

In the 1970s, P. A. Cundall developed a numerical method, namely the distinct element method (DEM), for computing the motion of large numbers of small objects [107, 108]. The DEM, which has since become one of the standard tools for numerical studies of system response in granular materials, enables researchers to predict the mechanical response of micro-granular systems of large numbers of particles through computer simulation.

On the other hand, scaling down the particles means that the traditional means of observing and identifying the configuration of a granular system does not apply to micro-granular systems. These experimental difficulties originate from the discrete nature of granular materials and their highly nonlinear inter-particle contact forces. This discreteness and the micro-scale sizes require efficient means to assemble the particles precisely, to excite them, and to measure their dynamic response. The high nonlinearity of these systems requires particularly high precision, and imperfections can be extremely important in controlling the dynamic response of the entire system.

Because of the difficulty of constructing repeatable micro-granular systems, the experimental study of micro-granular systems has been focused on unstructured granular systems, in which the exact configurations of micro-particles are unknown. The experimental studies on micro-granular systems are largely focused on the collective behavior of granular systems [109-111].

A special subset of ordered micro-granular systems is the micro-colloidal system, which consists of ordered two-/three-dimensional granular crystals that are created in water with self-assembly technology. Using photon correlation spectroscopy, Alan J. Hurd et al. measured the phonon dispersion curves and wavelength dependent friction factors [112]. More experimental studies using dynamic light scattering have also been performed to understand the over-damped collective behaviors; they have discussed both wall effects due to finite sample thickness (or confinement effects) and the role of ion behaviors in liquids [113, 114]. In 2004, P. Keim et al. first used video microscopy to study lattice dynamics, which allowed them to observe the harmonic lattice behavior of two-dimensional colloidal crystals with phonon dispersion curves [115, 116]. In later studies, lattice dynamics in one- and two-dimensional colloidal systems under various local substrate potentials produced by light have also been studied with video microscopy [117-120] and Brownian dynamics simulations [120, 121] to investigate the effects of local potential on the collective behaviors and phononic band structures.

Furthermore, the existence of fluid environments enriches the interaction forces by adding non-conservative hydrodynamic forces (such as viscous friction and many-body hydrodynamic forces) that arise from the relative motion of colloids [122-124]. Brownian dynamics simulation studies, where the motions are driven by entropic thermal fluctuations and the inertia of colloids is negligible due to their over-damped nature, have generally been performed to understand colloidal aggregation, phase transitions, and crystallization.

1.3. Contributions of this thesis

In this work, we study the fundamental dynamic response of micro-scale granular systems and design an innovative experimental platform that allows us to assemble, excite, and characterize ordered micro-granular lattices. This new experimental platform employs a laser system to deliver impulses with controlled momentum and non-contact measurements, including high-speed optical microscopy and laser interferometry, to detect the particles' displacement and velocity. We build and program a computer-controlled micro-manipulator that can position and assemble steel micro-particles in desired configurations for testing. We fabricate micro-structures to guide and confine the micro-particle assembly to allow interested dynamics to be tested. We test and demonstrate the capability of the laser excitation system to deliver controlled momentums to systems of dry (stainless steel particles of radius 150 µm) and wet (SiO_2 particles of radius 3.69 µm, immersed in fluid) micro-particles.

We first derive the governing equations of motion describing the dynamic response of dry and wet particles on a substrate, which we validate in experiments. To investigate the influence of micro-structure support on the dynamics of micro-particles loaded on the structure, we study the loss in our micro-particle configuration analytically and experimentally. We then measure the Stoke and Coulomb friction of the micro-particles by tracking particle-trajectories at varying initial momentum. Thereafter we study the collisions of rolling micro-particles in a groove to investigate the translational and angular momentum during collision. In observing inelastic collisions when the particles are rolling in the groove, we discover a linear dependency between the contact force and the tangential frictional force between the colliding particles. We also observe serious inelastic collisions when the spheres are rolling. Next we obtain an empirical equation of motions that describes the dynamics of the micro-granular system. We assemble one-dimensional dry chains of micro-particles and investigate the mechanical wave propagation properties as well as the influence of defects in these systems. Upon measuring the time of fly of wave that is propagating inside the chain at different initial input momentums, we show that measured group velocity depends on the initial velocity (which is a feature of wave traveling in granular systems with highly nonlinear interaction). We then examine the deviation of the measured group velocity with the Hertzian system and numerically show that the deviation can result from the presence of defects (which in this case are gaps between micro-particles). To prove this, we perform time of fly measurement for systems with a known maximum gap using the microscopic system and show that the measured group velocity agrees with numerical simulation.

We also study wave propagation in two-dimensional colloidal systems that are immersed in fluid. We employ self-assembling technology to create a two-dimensional hexagonal lattice in a micro-fluidic cell and apply the laser-based excitation to the system. We perform the experimental examination by sending laser energy into the system to excite the initial velocity of the six centermost particles in the lattice. The resulting high velocity (which is higher than what could be achieved using traditional means) allows us to explore the system while the particles have enough velocity to break through the hydrodynamic barriers and to study wave propagation in the system with different viscosities. We construct a model that includes the contact, hydrodynamic, electrostatic, and Stokes' drag forces and perform numerical simulation to study wave propagation at a time resolution higher than our experimental system. The simulation that results characterizes the roles of the hydrodynamic force and the contact within wave propagation and explains the origins of the isotropic wave propagation within the system.

Our finding represents the first systematic experimental and numerical analysis of wave propagation in ordered micro-granular systems. This work establishes the basis for further advancing studies of granular and colloidal systems and sheds light on the miniaturization of highly nonlinear granular devices.

1.4. Conceptual organization of this thesis

The remainder of the thesis is structured as follows:

In Chapter 2, we describe the design of the experimental platform that is used to carry out the micro-granular system.

In Chapter 3, we show the feasibility of utilizing pulsed laser ablation as a tool for delivering mechanical excitation to micro-particles. We experimentally calibrate the material response of the laser on different materials, as well as control the direction of transferred momentum.

In Chapter 4, we study the equation of motion of particles that are moving, rolling, sliding, and colliding in a groove. We also analyze the collisions between particles in a groove and provide empirical descriptions of the equation of motion for particles.

In Chapter 5, we construct one-dimensional micro-granular chains and study the wave-propagation within those chains. We measure the group velocity and attenuation of wave propagation. Via experiments and numerical simulation, we study the relationship between wave propagation and defects (the gap between micro-particles).

In Chapter 6, we apply the experimental setup to a micro-colloidal system and study the system's response to laser-generated impact. We also study the relation between striker velocity, viscosity, and decay length of the wave displacement of the particles in each force chain.