Crystallization of a quasi-two-dimensional granular fluid

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We experimentally investigate the crystallization of a uniformly heated quasi-2D granular fluid as a function of filling fraction. Our experimental results for the Lindemann melting criterion, the radial distribution function, the bond order parameter and the statistics of topological changes at the particle level are the same as those found in simulations of equilibrium hard disks. This direct mapping suggests that the study of equilibrium systems can be effectively applied to study non-equilibrium steady states like those found in our driven and dissipative granular system.

Equilibrium statistical mechanics is generally not applicable to systems far from equilibrium where both energy input and dissipation mechanisms are present, and identifying relevant tools for understanding these systems poses a serious challenge to the scientific community [1]. Granular materials have become a canonical system to explore such ideas since they are inherently dissipative due to inter-particle frictional contacts and inelastic collisions. Granular materials also have far reaching practical importance in a number of industries, but often accumulated ad-hoc knowledge is the only design tool used [2]. The dissipative nature of grains means that any dynamical study requires energy injection, typically involving vibration or shear [3]. An important feature of this class of systems is that the driving and dissipation mechanisms can be made to balance such that a steady state is achieved. Recent investigation of such Non-equilibrium Steady States have shown that connections with equilibrium statistical mechanics may provide an useful analogy. For example, a single particle on a turbulent air flow has been shown to exhibit equilibrium-like dynamics [4] and the nature of the melting phase transition in two-dimensional granular system is consistent with the KTHNY scenario for melting of equilibrium 2D crystals [5].

In our study we have developed an experimental system to generate a vibrated quasi-two-dimensional granular fluid of stainless steel spheres that is uniformly heated (i.e., energy injection is spatially homogeneous). In the insets (a) and (b) of Fig. 1 we present two such examples of typical non-equilibrium steady states for filling fractions $\phi = 0.60$ and $\phi = 0.76$, respectively. The first ($\phi = 0.60$) is a disordered dense fluid; there is a high collisional rate and at long times the particles randomly diffuse across the cell. The second ($\phi = 0.76$) is crystallized with each sphere packed into a hexagonal array locked by its six neighbors.

In this Letter we analyze the fluid-to-crystal transition as a function of filling fraction. The aim of our study is two-fold. Firstly, we make a quantitative characterization of the structural changes in the granular layer across this transition using a number of classic measures, namely the Lindemann criterion for melting, the radial distribution function, and the bond order parameter. Then we apply the novel concept of shape factor, recently introduced by Moucka and Nezbeda [6], to measure in detail the topology of the Voronoi cells across the crystallization transition. In parallel, we establish a direct comparison between the behavior of our experimental system and that of simulations of equilibrium hard disks and test the extent to which the above quantities, commonly used in equilibrium systems, can be used to study a non-equilibrium system such as ours.

Our experimental apparatus is adapted from a geom-

![Fig. 1: Lindemann ratio, $\gamma_m$, v.s. filling fraction, $\phi$, for a granular layer vibrated at $f = 50Hz$ and $\Gamma = 4$. The dotted horizontal line is located at $\gamma_m = 0.15$. Crystallization occurs at $\phi_s = 0.719$. Insets (a) and (b) are representative experimental frames in the fluid and crystal phases, at $\phi = 0.6$ and $\phi = 0.76$, respectively.](image_url)

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tery introduced by Olafsen and Urbach \[6\]. We inject energy into a collection of stainless steel spheres (diameter $D = 1.191 mm$) through sinusoidal vertical vibration with frequency $f$ and dimensionless acceleration, $\Gamma = A(2\pi f^2/g)$, where $A$ is the amplitude of vibration and $g$ is the gravitational acceleration. The spheres are confined in a fixed volume gap set by a horizontal stainless steel annulus (101.6 mm inner diameter) and sandwiched between two glass plates. The thickness of this annulus is $1.6 D$, thus constraining the system to be quasi-2D. The top glass plate is optically flat, but the bottom plate is roughened by sand-blasting generating random structures from $50 \mu m$ to $500 \mu m$. Upon vibration the rough plate homogeneously randomizes the trajectories of the particles. We record the dynamics of the system using high speed photography at $840 Hz$ and track the particle trajectories in a $(15 \times 15) mm^2$ central region.

The system is horizontal to minimize gravity induced effects such as rolling and compaction. We vary the total number of particles in the fixed volume cell over a wide range: from a single particle to an hexagonally packed crystal. We define the filling fraction of the granular layer as $\phi = N [D/(2R)]^2$, where $N$ is the total number of spheres, with diameter, $D$, in a cell of radius $R = 50.8 mm$. We fix the forcing parameters at $f = 50 Hz$ and $\Gamma = 4$ and systematically vary the filling fraction from $0.2 < \phi < 0.8$.

To interpret the qualitative change in behavior between dense fluid and crystalline phases, as $\phi$ is changed, we first measure the Lindemann ratio. For a wide range of materials, Lindemann found \[8\] that a solid melts when we first measure the Lindemann ratio. For a wide range of spheres, with diameter, $D$, and $\Gamma = 4$ and systematically vary the filling fraction $\phi$. The agreement between the experimental and numerical curves is remarkable, implying that our experimental non-equilibrium granular fluid is adopting structural configurations identical to those found in equilibrium hard disk systems. The only deviations occur near $r/D = 1$, as seen in the inset of Fig. 2(a) for $\phi = 0.60$. This discrepancy is due to the out of plane collisions in our experiments leading to apparent particle overlap in pro-

The Lindemann criterion is empirical and contains little information about the structural configuration. For this we calculate the radial distribution function $g(r)$, which is a standard way of describing the average structure of particulate systems \[10\]. In Fig. 2(a) we plot curves of $g(r)$ for representative $\phi$. For low filling fractions (e.g., $\phi = 0.5$) we observe fluid-like behavior, and $g(r)$ is peaked at $r/D = 1$, 2 and 3, as is commonly seen in hard sphere simulations \[10\]. At higher $\phi$ (e.g., $\phi = 0.65$), $g(r)$ develops an additional shoulder below the $r/D = 2$ peak, which at higher densities (e.g., $\phi = 0.7$ and $\phi = 0.72$) evolves into a distinct peak located at $r/D = \sqrt{3}$, signifying hexagonal packing. To each $g(r)$ experimental curve in Fig. 2(a), we have superposed a corresponding (dashed) curve from a Monte Carlo simulation of equilibrium hard disks recently reported by Moucka and Nezbeda \[8\], for identical values of $\phi$. The agreement between the experimental and numerical curves is remarkable, implying that our experimental non-equilibrium granular fluid is adopting structural configurations identical to those found in equilibrium hard disk systems. The only deviations occur near $r/D = 1$, as seen in the inset of Fig. 2(a) for $\phi = 0.60$. This discrepancy is due to the out of plane collisions in our experiments leading to apparent particle overlap in pro-

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FIG. 2: (a) Experimental (solid) and numerical (dashed, extracted from \[8\]) curves of the radial distribution functions for 5 values of $\phi$. The arrow points in the direction of decreasing $\phi$. Inset: Section of $g(r)$ curve for $\phi = 0.6$. (b) Radial distribution function at contact, $g(r = D)$ v.s. filling fraction. The dashed line corresponds to the theoretical Carnahan-Starling equation. $\phi_l$ and $\phi_s$ are the liquidus and solidus points, respectively.
ected phase, which would not be possible if the system were exactly two dimensional. The amount of overlap is consistent with our layer thickness of 1.6D. This deviation is seen in the plot of \( g(r = D) \) (i.e., at contact) which corresponds to the absolute maximum of \( g(r) \) and is shown in Fig. 2). For low filling fractions and up to \( \phi \sim 0.57 \), \( g(D) \) follows the theoretical curve of Carnahan-Starling, 

\[
g^{CS}(D) = \frac{16 - 7\phi}{16(1 - \phi)^2},
\]

which is usually assumed in the kinetic theory equation of state for granular gases \(^{11}\), but \( g(D) \) is systematically lower than \( g^{CS}(D) \) by \( \sim 1.14 \). For \( \phi > 0.57 \) the deviations from \( g^{CS}(D) \) increase up to \( \phi = 0.652 \) where there is a discontinuity in the curve’s slope. For \( 0.652 < \phi < 0.719 \) there is a period of slower growth of \( g(D) \) with \( \phi \). This is consistent with the scenario of the existence of a fluid phase (\( \phi < 0.652 \)), intermediate/transition phase (\( 0.652 < \phi < 0.719 \)) and crystal phase (\( \phi > 0.719 \)).

In addition to the development of correlations in the particle positions, angular correlations also arise as \( \phi \) is increased \(^{12}\). We measure these using the (global) bond-orientational order parameter \( \psi_6^{\text{global}} = \left| \frac{1}{M} \sum_{i=1}^{M} N_i \sum_{j=1}^{N_i} \exp(\theta_{ij}) \right| \), where \( M \) is the number of particles in the observation window, \( \theta_{ij} \) is the angle between the particles \( i \) and \( j \) and an arbitrary but fixed reference axis, and \( N_i \) is the number of nearest neighbors of particle \( i \), found using the Voronoi construction \(^{13}\). In Fig. 3 we plot the dependence of \( \psi_6^{\text{global}} \) on \( \phi \). The value of the bond orientational order parameter tends to unity in the crystal phase, but \( \psi_6^{\text{global}} \ll 1 \) for a disordered phase.

As with \( g(D) \), three different regions with the same phase boundaries: \( \phi_l = 0.652 \) (liquidus point) and \( \phi_s = 0.719 \) (solidus point) can be identified in Fig. 3 based on the slope of \( \psi_6(\phi) \). The observed behavior is consistent with the two-step continuous phase transition observed during equilibrium 2D crystallization \(^{12}\), where the first transition transforms the isotropic fluid phase into an hexatic phase with long range orientational ordering but no positional ordering and the second transforms the hexatic phase into a crystal with both long range orientational and positional order.

Moucka and Nezbeda \(^{12}\) have recently introduced the concept of shape factor, \( \zeta \), which is a sensitive measure to further quantify structural changes in the fluid-to-crystal transition in 2D. \( \zeta \) is defined at the particle level, by employing Voronoi tessellation, as \( \zeta_i = C_i^2/4\pi S_i \), where \( S_i \) is the surface area and \( C_i \) the perimeter of the Voronoi cell of the \( i \)th particle. For circles \( \zeta = 1 \) and \( \zeta > 1 \) for all other shapes (\( \zeta = 4/\pi \approx 1.273 \) for square, \( \zeta = \pi/5\tan(\pi/5) \approx 1.156 \) for regular pentagons, and \( \zeta = 6/(\sqrt{3}a^2) \approx 1.103 \) for regular hexagons). Therefore, \( \zeta \) is a quantifier of the topology of the Voronoi cells associated with the individual particles.

In Fig. 4(a) we present a surface plot of the distribution of shape factor, \( P(\zeta, \phi) \), and vertical cross-sections of \( P(\zeta, \phi) \) for fixed \( \phi \) are presented in Fig. 4(b). We superpose numerical (dashed lines) data of Monte Carlo of equilibrium hard disks \(^2\), for the same values of \( \phi \), and find that our experimental results are in excellent agreement with the numerical simulations. At low \( \phi \), \( P(\zeta) \) exhibits a broad and flat maximum; the particles are randomly distributed and no specific type of cells are formed. As \( \phi \) is increased, \( P(\zeta) \) becomes increasingly localized around the maximum which progressively moves towards lower values of \( \zeta \). Eventually, for \( \phi > 0.65 \) the distribution becomes bimodal and a distinct second maximum appears. In the vicinity of the crystallization point, \( \phi_s = 0.719 \), the original maximum for high \( \zeta \) values disappears while the low \( \zeta \) maximum rises sharply (centered at \( \zeta \approx 1.1 \), the value for regular hexagons). Fig. 4(a) clearly shows the existence of two distinct classes of shapes.

To quantify these classes we follow the classification scheme of the Voronoi cells proposed by Moucka and Nezbeda. An important point to note is that the location of the minimum of \( P(\zeta) \), where it exists, is only marginally dependent on \( \phi \), and we set \( \zeta_{\text{min}} = 1.159 \). Class \( A \) consists of particles with \( \zeta < \zeta_{\text{min}} \). Class \( B \) particles have \( \zeta_{\text{min}} < \zeta < \zeta_u \) and Class \( C \) have \( \zeta > \zeta_u \) where \( \zeta_u = 1.25 \). The upper bound, \( \zeta_u \), is set such that at the filling fraction for which both maxima of \( P(\zeta) \) have equal heights (\( \phi \approx 0.65 \)), the number of particles in classes \( A \) and \( B \) are the same. We plot the boundaries of cell classes on the surface plot of \( P(\zeta, \phi) \) in Fig. 4(a).

In Fig. 4(c) we present the \( \phi \)-dependence of the fraction of particles belonging to each of the Classes A, B and C. The nature of the previously mentioned special

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**FIG. 3:** Semi-logarithmic plot of the bond-orientational order parameter, \( \psi_6 \). The first two lines, I and II, are least squares fits of the form \( \psi \sim \exp[A\phi] \) and line III is a linear fit of the form \( \psi \sim A\phi \). The dashed and solid vertical lines are located at \( \phi_l = 0.652 \) and \( \phi_s = 0.719 \), respectively. Inset: Linear version of the plot.
FIG. 4: (a) Surface plot for the probability distribution functions of shape factor, $P(\zeta, \phi)$. The value PDF is given by the adjacent color bar. The two horizontal dashed lines located at $\zeta = 1.159$ and $\zeta = 1.25$ are the boundaries of classes A, B and C of the Voronoi cells, as defined in the text. (b) Experimental (solid) and numerical (dashed, extracted from [6]) vertical cross-sections of the $P(\zeta, \phi)$ distribution along 5 values of $\phi$. The arrow points in the direction of decreasing $\phi$. (c) Fraction of particles in the A, B and C classes, as defined in the text, as a function of filling fraction.

filling fraction values of $\phi_l$ and $\phi_s$, that separate the disordered liquid, the intermediate/transition phase and the crystal phases, becomes clear under this classification. $\phi_l = 0.652$ is the point at which Class A and Class B occurs in the same proportions (the fraction of Class C is negligible at this point). $\phi_s = 0.719$ is the point for which the fraction of Class B has sharply dropped to zero and the granular layer consists almost entirely of particle whose Voronoi cells are regular hexagons, i.e., crystallization occurs. It remains to be shown if the intermediate phase between $\phi_l$ and $\phi_s$ is simply a coexistence regions as suggested by the lever-like dependence of the fraction of Classes A and B, or this is an hexatic phase with algebraically decaying orientational order $^5$. One would need to perform the experiments with a considerably larger imaging window to have sufficient spacial extension to properly test such scalings.

In conclusion, we have reported detailed experimental measures of structural changes during the crystallization transition in a homogeneously heated granular fluid. Our results are in excellent quantitative agreement with Monte Carlo simulations for the crystallization of equilibrium hard disks. It is surprising that the particles in our granular layer adopt equilibrium-like structural configurations even though the system is both driven and dissipative, i.e., far from equilibrium. The equilibrium structural configurations for hard disks are usually determined by an entropy maximization argument $^{10}$. Whether we are able to explain the observed phase transitions in our system with entropic-like arguments similar to those used in hard sphere systems is an important question which arises from our study and needs further investigation.

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[1] D. A. Egolf, Science 287, 101 (2000).
[2] B. J. Ennis, J. Green, and R. Davies, Part. Technol. 90, 32 (1994).
[3] F. Melo, P. Umbanhowar, and H. L. Swinney, Phys. Rev. Let. 72, 172 (1994). B. Miller, C. O’Hern and R. P. Behringer, Phys. Rev. Let. 77, 3110 (1996).
[4] R. P. Ojha, P.-A. Lemieux, P. K. Dixon, A. J. Liu and D. J. Durian, Nature 427, 521 (2004).
[5] J. S. Olafsen and J. S. Urbach, Phys. Rev. Let. 95, 098002 (2005).
[6] F. Moucka and I. Nezbeda, Phys. Rev. Let. 94, 040601 (2005).
[7] J. S. Olafsen and J. S. Urbach, Phys. Rev. E 60, R2468 (1999). A. Prevost, D. A. Egolf and J. S. Urbach, Phys. Rev. Lett. 89, 084301 (2002).
[8] F. A. Lindemann, Phys. Z. 11, 609 (1910).
[9] B. J. Alder and T. E. Wainwright, Phys. Rev. B 19, 2457 (1979). A. Jaster, Phys. Rev. E 59, 2594 (1999).
[10] D. P. Fraser, M. J. Zuckermann and O. G. Mouritsen, Phys. Rev. A 42, 3186 (1990).
[11] H. Kawamura, Prog. Theor. Phys. 61, 1584 (1979).