Non-equilibrium steady states, coexistence and criticality in driven quasi 2D granular matter

Thomas Schindler† and Sebastian C. Kapfer†

Theoretische Physik 1, FAU Erlangen-Nürnberg, Staudtstr. 7, 91058 Erlangen, Germany

(Dated: November 14, 2018)

Non-equilibrium steady states of vibrated inelastic frictionless spheres are investigated in quasi-two-dimensional confinement via molecular dynamics simulations. The phase diagram in the density-amplitude plane exhibits a fluid-like disordered and an ordered phase with threefold symmetry, as well as phase coexistence between the two. Moreover, there is a square bilayer state which is connected to the fluid by BKTHNY-type two-step melting with an intermediate tetratic phase. The critical behavior of the two continuous transitions is studied in detail. For the fluid-tetratic transition critical exponents of $\tilde{\gamma} = 1.73$, $\eta_4 = 0.25$, and $z = 2.05$ are obtained. The phase diagram topology is incompatible with any equilibrium free energy and features an anomalously diluted fluid in coexistence with the threefold cluster. A dynamical mechanism exists that brings about metastable traveling clusters and at the same time stable clusters with anisotropic shapes at low vibration amplitude.

PACS numbers: 45.70.Mg, 64.75.St, 05.70.Jk

I. INTRODUCTION

The slab geometry of vibrating plates filled with inelastic granular spheres is a particularly interesting setup in the field of non-equilibrium statistical physics. The macroscopic particles (typical size $\sim 1 \text{ mm}$) dissipate kinetic energy at each collision. Fluidized states can be maintained by energy input to the vertical particle motion via the vibrating plates. This energy is then partly transferred to horizontal motion in particle-particle collisions. The energy flow in this injection, transfer, and dissipation mechanism breaks detailed balance and the system is inherently strongly out of equilibrium. The non-equilibrium property manifests itself in several interesting phenomena, which have been investigated largely in the past two decades. Among these are inelastic collapse, heterogeneous granular temperatures, non-Gaussian velocity distributions, segregation of mixtures, the Kovacs memory effect, and inelastic hydrodynamic modes.

Two-dimensional (2D) driven granular matter excels as a model system for non-equilibrium statistical mechanics for several reasons. Particle trajectories are comfortably accessible in experiments by filming from the top. Moreover, the influence of gravity is tunable via the choice of the driving amplitude and frequency. Finally, the particles can be agitated homogeneously throughout the horizontal directions. Any in-plane inhomogeneity thus emerges from spontaneous symmetry breaking.

After some relaxation time, a non-equilibrium steady state (NESS) is reached in which the energy injection, transfer, and dissipation rates balance. Because of their non-equilibrium nature, the involved phases in these states are no thermodynamic phases in the strict sense. Even though these phases show intriguing resemblance to the corresponding thermal equilibrium system of colloidal particles, the NESS usually retain residual energy and particle flows which cannot occur in equilibrium.

The phase behavior depends decisively on parameters such as box dimensions and roughness, filling density, driving frequency and amplitude, and inelasticity of the particles. Several studies have investigated the phase behavior as functions of different control parameters. A complete phase diagram in the multi-dimensional parameter space, however, is not at hand. For the parameters studied here, the NESS comprise isotropic fluid-like phases as well as square and hexagonal monolayers and bilayers. In equilibrium, the transitions between those phases are all of first-order type. By contrast, in experiments with the shaken granular particles, a continuous transition between an isotropic phase and a phase with square order has been reported, with diverging correlation functions and several critical exponents measured. In molecular dynamics (MD) computer simulations the same phases were found, but divergences of the correlation functions were not reproduced.

The fact that – despite extensive studies – the picture of the quasi-2D vibrated granulates is still incomplete led us to revisit the system with MD simulations. We employ an altered approach to refine the description of the continuous fluid-square transition and characterize it as BKTHNY-type two-step transition with an intermediate tetratic phase, which is reported for the first time. Moreover novel non-equilibrium phenomena are described, namely an anomalous dilution in the fluid-threefold coexistence and emergent particle currents at the surfaces of threefold clusters.

The paper is organized as follows. In Sec. III we present the system and model and give technical details about the simulation and the order parameter. Secs. IV-VI summa-
rize our main findings on the NESS. Firstly we discuss the relevant parameters for the formation of ordered phases and sketch the NESS state diagram in Sec. III. Secondly, the fluid-square transition contained in the phase diagram is thoroughly characterized in Sec. IV including precise values for the critical exponents. Finally, the mechanism that brings about anisotropic shape and persistent motion of threefold clusters is discussed in Sec. V.

In Sec. VI we conclude by comparing the phase behavior to previous studies and discuss the non-equilibrium nature of the observed effects.

II. SYSTEM SETUP AND PARAMETERS

Computer simulations are performed in a shallow cuboidal box of spatial dimensions $L \times L \times h$ with periodic boundary conditions applied in $x$ and $y$ directions. In $z$ direction the box is confined by two hard walls. The space in between the walls contains $N$ hard spheres of diameter $\sigma$ at a projected number density $\rho \equiv N/L^2$. Gravitational acceleration $g$ acts on the particles in negative $z$ direction implying a timescale $\tau_0 \equiv \sqrt{\sigma/g}$. The walls oscillate in-phase with displacement $A \sin(\omega t)$ in $z$ direction, where $A$ is the driving amplitude, $\omega$ is the angular frequency of the driving and $t$ denotes time. All simulations in this work are carried out with $h = 1.83\sigma$ and $\omega\tau_0 = 12$. The height was chosen such that a continuous transition from fluid to square could be observed. The value of $\omega$ lies within the high-frequency regime, where the nucleation of the threefold cluster upon increasing $A$ is insensitive to $\omega$.

The trajectories of the particles are calculated by an event-driven MD algorithm [26]. Particle-particle and particle-wall collisions are modeled as inelastic instantaneous collisions [27] with a constant coefficient of restitution $e = 0.95$ for the momentum transfer normal to the surfaces (where $e = 1$ and $e = 0$ would correspond to elastic and fully inelastic collisions, respectively). The model conserves momentum in $x$ and $y$ directions (momentum in $z$ direction changes in particle-wall collisions). We do not include transfer of momentum tangential to the particle surfaces, i.e., the Coulomb friction coefficient vanishes. Hence there is no coupling of rotational and translational degrees of freedom. Our model thus depends only on a single parameter—the coefficient of restitution—yet still features energy injection, transfer, and dissipation mechanisms. Despite this simplification, our phase diagram qualitatively agrees with earlier studies that do include the rotational degrees of freedom.

Most of our analysis and the color coding of all snapshots in this work are based on the (projected 2D) fourfold local bond-orientational order parameter $\psi_4^{(n)}$ of a particle $n$. We use a refined version [28],

$$\psi_4^{(n)} \equiv \sum_{m=1}^{N_n} \frac{w_{nm}}{W_n} e^{i\alpha_{nm}},$$  \hspace{1cm} (1)

with weight factors differing from the usual definition. Here the sum is carried out over the $N_n$ Voronoi nearest neighbors of particle $n$, the weight factor $w_{nm}/W_n$ is the length $w_{nm}$ of the Voronoi facet shared by particles $n$ and $m$ (marked in green in Fig. 1) normalized by the total circumference $W_n$ of the Voronoi cell of particle $n$ (green plus blue lines), and $\alpha_{nm}$ (red) is the angle between the $x$ axis and the connection line of particles $n$ and $m$. The weight factors are included to make the order parameter a continuous function of particle positions and in particular robust against small distortions in lattices by minimizing the influence of e.g. diagonal nearest neighbors in distorted square lattices (like the top left particle in Fig. 1). The modulus of $\psi_4^{(n)}$ ranges from 0 for particles with no local fourfold symmetry to 1 for particles centered in a square of four nearest neighbors.

Unless otherwise stated, each simulation run is started from a special configuration prepared to minimize nucleation effects. The configuration consists of two domains of square and threefold order, respectively, immersed in a fluid. (The phases are described in detail in Sec. III.) Velocities are initialized from Gaussian distributions, where the mean velocity of the $N$ particles is subtracted yielding zero net velocity. After a relaxation period the simulations reach steady states which are the subjects of our investigations. We explicitly verified that all steady states are stable when the simulation is paused and the velocities are reset to a Gaussian distribution.

III. PHASE DIAGRAM

Various aspects of the phase diagram have been reported previously [11, 17, 22], but a complete picture is lacking so far. In Fig. 2(a) we present the phase diagram in the $\rho-A$ plane, obtained for $N = 4000$ particles. (The transition lines shown here are shifted with respect to the true ones by finite-size effects.) For the chosen parameters, the system exhibits both transitions
FIG. 2. (color online) (a) Phase diagram in the density-amplitude plane for box height $h = 1.83\sigma$ and angular frequency $\omega\tau_0 = 12$. Investigated state points are on a grid with spacings $\Delta\rho\sigma^2 = 0.02$ and $\Delta A = 0.002\sigma$. The simulation at each state point was carried out with $N = 4000$ particles and averaged over a time interval $t = 5000\tau_0$. The maximum density $\rho_{\text{max}}\sigma^2 \approx 2.31$ is slightly larger than the density of two hexagonal close packed layers, $\rho\sigma^2 = 4/\sqrt{3}$ due to the possibility of buckling. Below the blue line the fluidized state collapses and all particles drop to the bottom plate. The green line indicates evaporation of a threefold cluster upon decreasing $\rho$. Between $0.02\sigma \leq \rho \leq 0.04\sigma$ the nucleation density (gray line) of the threefold cluster upon compression differs from the evaporation density, with a hysteresis region (hatched) between the green and gray lines. The purple line marks the density where the threefold cluster comprises the whole simulation box. The red and yellow lines indicate the continuous fluid-tetratic and tetratic-square transitions, respectively. Labels (c)–(j) relate the state points to the snapshots in the following panels. The inset shows a wider view of the phase diagram up to amplitude $A = 1\sigma$.

(b) Sketch of the color coding of the particles in snapshots (c)–(j). The hue is determined by the complex phase $\arg(\psi_4^{(n)})$ of the fourfold order parameter. The color saturation indicates the modulus of $\psi_4^{(n)}$ ($|\psi_4^{(n)}| = 0$ → gray, $|\psi_4^{(n)}| = 1$ → fully saturated). 

(c) $\rho\sigma^2 = 1.2$, $A = 0.004\sigma$  
(d) $\rho\sigma^2 = 1.0$, $A = 0.03\sigma$  
(e) $\rho\sigma^2 = 1.2$, $A = 0.1\sigma$  
(f) $\rho\sigma^2 = 1.46$, $A = 0.03\sigma$  
(g) $\rho\sigma^2 = 1.5$, $A = 0.03\sigma$  
(h) $\rho\sigma^2 = 1.7$, $A = 0.03\sigma$  
(i) $\rho\sigma^2 = 1.7$, $A = 0.05\sigma$  
(j) $\rho\sigma^2 = 1.7$, $A = 0.03\sigma$

Fluid threefold coexistence (circular cluster shape).  
Fluid threefold coexistence (partially wetted).  
Fluid-threefold coexistence with cluster percolating the box in $y$ direction [metastable; same state point as (h)]. $\varphi$ denotes the angle between the $y$ axis and the symmetry axis of the lattice unit cell (see Sec. [V]). The big arrow indicates the direction of motion of the cluster.
with first-order character as well as critical behavior.

At low driving amplitudes \( A < 0.007 \sigma \), we observe inelastic collapse \[27\] in which all spheres drop to the bottom plate because the injected energy does not suffice to maintain a fluidized state. A snapshot of this state is displayed in Fig. 2(c); as in all snapshots, the color code shows the local orientation of the square order, i.e., the complex phase of the \( \psi_4 \) order parameter as depicted in Fig. 2(h).

At higher \( A \) there are several fluidized states described in the following. At low \( \rho \), the system is in a homogeneous unordered fluid phase, exemplarily seen in Fig. 2(d). In the high-density limit, we find a lattice with threefold symmetry. This lattice consists of two hexagonal layers offset against each other, such that particles of the top layer sit in the dips of the bottom layer, as in the hexagonal close-packed structure. In projection, one finds a honeycomb lattice with three nearest neighbors. Because bottom and top layer particles are distinct in the gravitational field, this lattice only has threefold rotational symmetry and not sixfold as the honeycomb (see also Sec. V). The transition from the fluid to the threefold lattice is different in the high-amplitude regime \( A \gtrsim 0.05 \sigma \) and in the moderate-amplitude regime \( 0.007 \sigma < A \lesssim 0.05 \sigma \).

In the high-amplitude regime the transition exhibits the phenomenology of a first-order phase transition. At the nucleation density \( \rho_0 \sigma^2 \approx 0.9 \), a cluster with threefold structure emerges [see Fig. 2(e)] which then grows with increasing \( \rho \) until it comprises the entire box for \( \rho \sigma^2 \gtrsim 2.26 \). This transition scenario is stable up to at least \( A = 1 \sigma \) as shown in the inset of Fig. 2(a). The relatively broad coexistence region as compared to the thermal equilibrium system \[19\] is an effect of enhanced dissipation in the dense phase and has been reported previously \[5, 17, 22\].

In the moderate-amplitude regime, the fluid contains patches with square bilayer structure Fig. 2(f) for \( \rho \sigma^2 \gtrsim 1.4 \). The length scale and life time of these patches (as seen in movie #1 in the supplement \[29\]) diverges upon increasing \( \rho \), and the system undergoes a continuous transition. The result is the tetratic state as seen in Fig. 2(g). This phase is distinguished from a true solid by the presence of dislocations at which grid lines end [marked in Fig. 2(g)]. In a second continuous transition at higher \( \rho \), the density of dislocations vanishes and a square bilayer solid is formed (no snapshot shown). This two-step transition hence displays the phenomenology of the BKTHNY theory \[30, 31\] and is analyzed in greater detail in section IV. In contrast to the large density difference between the fluid and the threefold phase, we do not find any evidence of density inhomogeneities at the fluid-tetratic or tetratic-square transitions.

At higher \( \rho \), a first-order-type transition from the square phase to the threefold lattice is found with an evaporation density of the threefold cluster of \( \rho \sigma^2 = 1.57 \). Surprisingly, in the coexistence region, the square phase is destabilized by the presence of the threefold cluster and melts into a fluid, see Fig. 2(h). Strikingly, this fluid has a lower density than the evaporation density of the threefold cluster. This discrepancy is a genuine non-equilibrium effect (see conclusion). The anomalous dilution of the fluid is not a finite-size effect and persists in simulations with larger \( L \), see movie #2 in the supplement \[29\].

To verify that the fluid-threefold coexistence is the true NESS above the nucleation density, additional simulations were performed in that region initialized as pure square phases (including grain boundaries to facilitate nucleation) or fluids. In these, one observes a very low nucleation rate of threefold clusters. In the range \( 0.02 \sigma \leq A \leq 0.04 \sigma \) and at \( \rho \sigma^2 \lesssim 1.71 \), however, we could not observe any nucleation events at all, which is indicated in the phase diagram as a hatched hysteresis region. As reason for this hysteresis we identify the incompatible symmetries of the square and threefold phases. The metastable states – i.e., before nucleation of a threefold cluster – exhibit fluid-fluid coexistence in the range \( 0.05 \sigma \leq A \leq 0.068 \sigma \) and fluid-square coexistence for \( A \geq 0.068 \sigma \), qualitatively consistent with the findings of Guzmán et al. \[24\].

Finally we discuss the shape of the threefold cluster which appears in the fluid-threefold coexistence. For large \( A \), the shape of the cluster is dominated by an isotropic surface tension and thus is close to circular, Fig. 2(e). At lower \( A \) the symmetry breaking between the top and bottom hexagonal layers due to gravity is enhanced. This leads to a pronounced anisotropy of the surface tension, and to the emergence of three stable and three unstable directions in the hexagonal bilayer (see Sec. V). The three stable facets grow out to become the three corners of a cluster with triangular shape, see Figs. 2(h) and (i). The remaining interfaces of this cluster are of the unstable facet type.

With suitable initialization one can also obtain a metastable state with a stripe-shaped threefold cluster stabilized by the periodic boundary conditions Fig. 2(j). If the two fluid-solid interfaces are inequivalent, the cluster may absorb new particles on one interface [right-hand side in Fig. 2(j)], and dissolves on the other (left), leading to the curious effect that the ordered domain is effectively propelled forward, see movie #3 in the supplement \[29\]. The mechanism behind this and the dependence of the effective cluster speed on its orientation and \( A \) is examined in Sec. V.

IV. FLUID-TETRATIC-SQUARE TRANSITION

The freezing of the fluid to the square solid proceeds in two continuous phase transitions at two different critical densities. At the lower density, the fluid transforms into a tetratic state by divergence of the length and time scales of ordered patches. In the following, we measure the emerging orientational order and calculate precise values for the fluid-tetratic critical density \( \rho_0 \) and the critical exponents governing the divergences. The system is driven
through the transition by increasing $\rho$ at several fixed $A$.

At first the global orientational order for several system sizes, ranging from $L = 40\sigma$ to $160\sigma$, and $A = 0.03\sigma$ is considered as an indicator for a continuous transition. The degree of global orientation is measured via the order parameter

$$\Psi_4 \equiv \left\langle \frac{1}{N} \sum_{n=1}^{N} \psi_4^{(n)} \right\rangle. \tag{2}$$

The typical behavior of a continuous transition is observed (cf. Fig. 3). In the fluid phase – like the one displayed in Fig. 2(d) – contributions of the differently oriented particles cancel out yielding zero mean. When approaching the critical point patches with fourfold orientational order emerge, which increase in size and eventually reach the scale of the box. In this region there are only a few patches [see e.g. Fig. 2(f)] and their contributions will not cancel completely but yield a finite average. The effect is more prominent with smaller simulation boxes and sets in at lower $\rho$. When further increasing $\rho$, there is only one domain left and the whole system orders yielding a strong increase of $\Psi_4$. The resulting kink in the data is at a density lower than $\rho_4$ but it approaches $\rho_4$ in the limit of $L \to \infty$. Note that because the tetratic phase does not exhibit true long-range orientational order, an exponent $\beta$ governing the order parameter via $\Psi \sim (-\epsilon)^\beta$, does not exist in the infinite system.

Length and time scales are studied via the (fourfold)

intermediate scattering function $^{33}$

$$F_4(k, \tau) \equiv \frac{1}{N} \left\langle \sum_{m=1}^{N} \sum_{n=1}^{N} \mathrm{e}^{i k \cdot (r_m(t) - r_n(t + \tau))} \right\rangle,$$  \tag{3}

where $k$ is a 2D wave vector, $r_m$ is the 2D projection of the position of particle $m$, $\tau$ is a time difference, the bar denotes complex conjugation, and the angle brackets denote time average. At equal times, $F_4$ is the static structure factor, $S_4(k) \equiv F_4(k, 0)$.

In the fluid, for $\rho < \rho_4$, Ornstein-Zernikе behavior $^{33}$

$$S_4(k) = \frac{\lambda_4}{1 + (\xi_4 k)^2}, \tag{4}$$

at low wave numbers $k \equiv |k|$ is observed. We determine the static susceptibility $\chi_4$ and the orientational correlation length $\xi_4$ \[i.e., the typical size of a patch as seen in Fig. 2(f)] by fitting our simulation data to Eq. (4). Figure 3(a) shows $S_4(k)$ for several densities and $L = 120\sigma$ exemplarily at $A = 0.03\sigma$. The data for other values of $A$ show qualitatively the same behavior. The extracted values of $\xi_4$ are displayed in Fig. 4(b). The data exhibits divergence at $\rho_4$ of the XY model type $^{34}$,

$$\xi_4 \propto \exp(b/\sqrt{\epsilon}), \tag{5}$$

with constant $b$ and reduced density parameter $\epsilon \equiv 1 - \rho/\rho_4$. As the finite system already orders at lower $\rho$, fits are restricted to the region below the apparent maximum of $\xi_4$ (red data points). The divergence of $\chi_4$ is closely linked to the divergence of $\xi_4$, as shown in Fig. 4(b). One observes power law dependence

$$\chi_4 \propto \xi_4^{\tilde{\gamma}}, \tag{6}$$

with critical exponent $\tilde{\gamma}$. The resulting values of $b$, $\rho_4$, and $\tilde{\gamma}$ are displayed together with values obtained for other $A$ at $L = 80\sigma$ in Tab. I.

The critical slowing down of large patches is quantified by measuring the correlation time $\tau_4$ and the dynamic critical exponent $z$ in the limit of low $k$. After an initial decay of all but the slowest mode, the long-time asymptotics of the intermediate scattering function $F_4$ has an exponential tail,

$$F_4(k \to 0, \tau) = C_\tau \exp(-\tau/\tau_4), \tag{7}$$

with the constant prefactor $C_\tau \propto \chi_4$ due to the initial decay. The simulation results for $L = 120\sigma$ and fits are displayed in Fig. 5(a) exemplarily at $A = 0.03\sigma$. To extract the asymptotic exponential decay, we fit data for $\tau \geq 50\tau_0$. The results for $\tau_4$ are shown in Fig. 5(b) as functions of $\xi_4$. Again $\tau_4$ diverges with $\xi_4$ as

$$\tau_4 \propto \xi_4^{\tilde{\gamma}}, \tag{8}$$
which defines $z$ (also shown in Table I). Assuming that the values are constant along the critical line, the best estimates averaging over the values for different $A$ are

$$\hat{\gamma} = 1.73 \pm 0.07, \quad z = 2.05 \pm 0.06. \quad (9)$$

Finally the fourfold correlation function $g_4(r)$ is examined to estimate the anomalous dimension $\eta_4$. We define

$$g_4(r) \equiv \frac{1}{\rho N^4} \left\langle \sum_{m=1}^{N} \sum_{n=1}^{N} \delta(r + r_m(t) - r_n(t)) \right. \times \left. \phi_4^{(m)}(t) \phi_4^{(n)}(t) \right\rangle, \quad (10)$$

which is in practice calculated via backwards Fourier transforming $S_4(k)$. Plotting $g_4$ on double logarithmic scale (see Fig. 5) for box size $L = 160\sigma$, one can distinguish between short-range exponential decay for $\rho \sigma^2 \lesssim 1.465$ and quasi-long-range algebraic decay $g_4 \propto r^{-\eta_4}$ for $\rho \sigma^2 \gtrsim 1.47$. The algebraic decay at the transition is well described by a power law with anomalous dimension

$$\eta_4 = 1/4. \quad (11)$$

Now we turn to the tetratic-square transition, at which the density of free dislocations vanishes. The length scale associated with this density can be extracted from the decay of the two-dimensional pair correlation function $g(x, y)$ towards 1. To reduce noise, multiple samples are averaged coherently [35, 36], i.e., they are aligned such that their individual global orientational order parameter $\Psi_4$ is real and positive. (Note that this procedure is necessary rather than investigating the radial distribution function $g(r)$, where the azimuth $\phi$ has been averaged over. Rapid decay of $g(r) - 1$ would be insufficient to demonstrate short-range positional order, as $g(r) - 1$ decays rapidly even in a solid. The same is true for a naive average, where multiple configurations with different $\Psi_4$ orientations are incoherently averaged over.)

Figure 7(a) shows the (coherent) direct correlation function $h(x) \equiv g(x, 0) - 1$ at some exemplary densities for box size $L = 120\sigma$. The system exhibits short-range exponential decay of the envelope,

$$h(x) \propto \exp(-x/\xi_{\text{pos}}) \quad \text{for} \quad \rho \sigma^2 \lesssim 1.54, \quad (12)$$

where $\xi_{\text{pos}}$ is the positional correlation length, i.e., the typical distance of dislocations. The inset shows the fitted values of $\xi_{\text{pos}}$ for all obtained exponentials. With

| $A/\sigma$ | $\rho_4 \propto \exp(b/\sqrt{\varepsilon})$ | $\chi_4 \propto \xi_4^\gamma$ | $\tau_4 \propto \xi_4^z$ |
|-----------|-------------|----------------|---------------|
| 0.01      | 1.62 ± 0.03 | 0.8 ± 0.4     | 1.62 ± 0.21  |
| 0.014     | 1.56 ± 0.02 | 0.70 ± 0.15   | 1.73 ± 0.09  |
| 0.02      | 1.52 ± 0.01 | 0.74 ± 0.14   | 1.76 ± 0.15  |
| 0.03      | 1.51 ± 0.01 | 0.74 ± 0.05   | 1.79 ± 0.16  |
| 0.04      | 1.55 ± 0.03 | 1.3 ± 0.4     | 1.8 ± 0.4    |

TABLE I. Critical densities $\rho_4$, parameter $b$, and critical exponents $\gamma$ and $z$ of the fluid-tetratic transition, for several amplitudes $A$ obtained from fitting Eqs. (5), (6), and (8).
increasing \( \rho \), \( \xi_{pos} \) increases implying a decrease of the number of dislocations. Ultimately the system crosses over to algebraic quasi-long-range behavior

\[
h(x) \propto x^{-\eta_{pos}} \quad \text{for} \quad \rho \sigma^2 \geq 1.54,
\]

with \( \eta_{pos} \) being the anomalous dimension of positional order. The two types of asymptotics are distinguished by plotting \( h(x) \) on double logarithmic scale as done for the envelope in Fig. 5(b). At the crossover density \( \rho \sigma^2 = 1.54 \) the average number of dislocations in the simulation box is of the order of 1. Therefore, one observes switching between short-range and quasi-long-range behavior [as indicated by the two curves shown in Fig. 5(b)] at very long time scales \( \geq 10^4 \tau_0 \) depending on the number of dislocations at the respective instant of time. From the average over times at which quasi-long-range behavior is exhibited, we infer a value of \( \eta_{pos} \approx 1/3 \). The critical density \( \rho_{pos} \), where dislocations become infinitely sparse

\[
F_4(\tau) \sim C_4 e^{-\tau/\tau_4}
\]

at very long times. The two types of asymptotics are distinguished by \( \tau_4 \geq 10^{-1} \) depending on the number of dislocations at the respective instant of time. From the average over times at which quasi-long-range behavior is exhibited, we infer a value of \( \eta_{pos} \approx 1/3 \). The critical density \( \rho_{pos} \), where dislocations become infinitely sparse

\[
F_4(\tau) \sim C_4 e^{-\tau/\tau_4}
\]

at very long times. The two types of asymptotics are distinguished by \( \tau_4 \geq 10^{-1} \) depending on the number of dislocations at the respective instant of time. From the average over times at which quasi-long-range behavior is exhibited, we infer a value of \( \eta_{pos} \approx 1/3 \). The critical density \( \rho_{pos} \), where dislocations become infinitely sparse

\[
F_4(\tau) \sim C_4 e^{-\tau/\tau_4}
\]

at very long times. The two types of asymptotics are distinguished by \( \tau_4 \geq 10^{-1} \) depending on the number of dislocations at the respective instant of time. From the average over times at which quasi-long-range behavior is exhibited, we infer a value of \( \eta_{pos} \approx 1/3 \). The critical density \( \rho_{pos} \), where dislocations become infinitely sparse

\[
F_4(\tau) \sim C_4 e^{-\tau/\tau_4}
\]
FIG. 7. (color online) (a) Scan through the two-dimensional direct correlation function $h$ along the direction $x$ of the order parameter $\Psi_L$ at some exemplary densities as indicated and box size $L = 120\sigma$. Multiple samples have been aligned and averaged over coherently, see main text. The black straight lines show exponential fits of the maxima of $h(x)$ to Eq. (12). The inset shows the resulting fit values of the positional correlation length $\xi_{\text{pos}}$ for all obtained exponentials. (b) Maxima of the data shown in panel (a) on double logarithmic scale for a wider range of densities from $\rho\sigma^2 = 1.46$ to 1.70. For $\rho\sigma^2 = 1.54$ the times at which the system exhibits exponential (dashed curve) or algebraic (dashed-dotted curve) decay, were averaged separately. The black straight line corresponding to a power law with exponent $-1/3$ is a guide to the eye.

![Graph of direct correlation function](image)

![Graph of direct correlation function](image)

![Graph of direct correlation function](image)

![Graph of direct correlation function](image)

![Graph of direct correlation function](image)

![Graph of direct correlation function](image)

FIG. 8. (color online) (a) Top view sketch of the threefold lattice consisting of two hexagonal layers. The rhombus in the center represents a unit cell of the lattice with its long diagonal being one of the three symmetry axes (dashed-dotted lines). The orientation of a cluster is measured by the angle $\varphi$ between this diagonal and the $y$ axis of the box. Arrows at the edge show how top layer particles are supported by bottom layer particles from the exterior at the stable and unstable facets, respectively, and hence illustrate the asymmetry between these facets. In the lattice displayed here $\varphi = 0^\circ$. This orientation has two equivalent left and right interfaces in configurations with interfaces parallel to the $y$ direction [as in Fig. 2(j)], whereas a lattice rotated by $\varphi = 30^\circ$ would constitute the maximally asymmetric case with a stable facet to the right and an unstable facet to the left. The case $\varphi = 60^\circ$ is the same as $\varphi = 0^\circ$ mirror-inverted in $y$ direction and therefore again symmetric with respect to the $y$ axis. Therefore it is sufficient to consider angles between $0^\circ$ and $30^\circ$. (b) Sketch of the calculation of the $x$ coordinate of the center of mass, $x_{\text{com}}$, with periodic boundary conditions via mapping of the $x$ coordinates of the particles onto the unit circle in the complex plane. Particles are displayed as red circles, the average of the mapped coordinate is depicted as blue circle. Symbols are declared in the text.

![Sketch of threefold lattice](image)

![Sketch of threefold lattice](image)

![Sketch of threefold lattice](image)

![Sketch of threefold lattice](image)

![Sketch of threefold lattice](image)

serve momentum in the $x$ and $y$ direction. Indeed, the mean velocity of the particles due to rounding errors in the simulations is lower than $10^{-10}\sigma/\tau_0$ throughout the simulations and is therefore not accountable for the measured directed cluster motion.

The center of mass motions for different cluster orientations $0^\circ \leq \varphi \leq 30^\circ$ are shown in Fig. 9(a). From these the average cluster speeds are calculated by measuring the end-to-end distance and dividing by the time interval [see Fig. 9(b)]. As expected, one observes zero cluster speed for symmetric interfaces, i.e., when left and right interface both are half-way between the stable and unstable facets. For non-zero angles, the cluster starts to move in the direction where the interface is composed predominantly of the stable facet. The cluster speed increases with increasing asymmetry between the interfaces. For angles greater than $20^\circ$ the speed decreases again, although the cluster becomes even more asymmetric here.

The mechanism of the advancing cluster relies on particle exchange with wetting films of square symmetry on its boundary. Particles are absorbed in a zipper-like fashion at the front interface, and detach from the rear interface. The effect of this process is a translation of the cluster in the positive $x$ direction, see movie #4 in the supplement. When approaching $\varphi = 30^\circ$, however, the right-hand interface becomes parallel to the stable facet, and lacks the kinks necessary for the zipper mechanism to function. This explains the decrease of the cluster speed at these angles.

We also measure the cluster speed as a function of $A$ as shown in Figs. 9(c) and (d). Here one can see that the cluster speed decreases with increasing $A$. This is paralleled by the clusters becoming more circular in the non-percolated configurations [cf. Figs. 2(e), (h), (i)]

![Sketch of cluster motion](image)

![Sketch of cluster motion](image)

![Sketch of cluster motion](image)

![Sketch of cluster motion](image)

![Sketch of cluster motion](image)
and hence confirms that the mechanism that leads to the faceted cluster shape is the same as the one driving the cluster propulsion. Two reasons for the decreased cluster speed are identified. Firstly, the square film is thinner at higher $A$ and therefore the zipper mechanism does not work as efficiently. Secondly, the decreasing influence of gravity as compared to the driving reduces the asymmetry between the two types of facets.

VI. DISCUSSION AND CONCLUSION

The phase diagram of the vibrated quasi 2D granular sphere system exhibits a first-order transition to a threefold lattice as well as a continuous fluid-square transition with intermediate tetratic phase. The densities on the fluid and threefold side of the first-order transition are consistent with values reported for simulations at $A = 0.15\sigma$ by Melby et al. [17]. The critical amplitude for the inelastic collapse — best compared in terms of the dimensionless acceleration $\Gamma = A\omega^2/g = 1.01 \pm 0.14$ — is also in fair agreement with previous studies, see e.g. [1, 2]. All of these previous studies involve tangential friction. Therefore, the consistency with our results demonstrates that tangential friction is not essential for the phase behavior in fluidized granulates.

The fourfold ordering transition found in experiments [24, 25] and simulations [23] is consistent with the topology of the phase diagram presented here. Quantitative agreement between simulation and experiment is not expected due to the subtle role of roughness of particle surfaces [23]. Our study departs from the earlier studies in two ways. First, instead of the driving amplitude $A$, we control the transition via the global density $\rho$; second, the earlier studies examine configurations with a fluid-fluid phase separation, in which the denser of the two fluids undergoes an ordering transition by increasing $A$. Due to the phase separation, the density of the fluid is not strictly fixed either. For our set of parameters, there is no stable fluid-fluid phase separation which considerably simplifies the analysis and permits to directly control the density. Therefore, our approach yields the
critical exponents associated with the control parameter $\rho$, without admixture of $A$. We expect that the tetratic phase is also observable in experiment. The positional correlation length is $\sim 3$ particles when the tetratic first forms, which indicates that short-range order will be detectable even in very small systems.

For the fluid-tetratic transition, precise values for $b$ and $A_4$ were calculated. We precisely measured the critical exponents $\tilde{\gamma} = 1.73 \pm 0.07$ and $z = 2.05 \pm 0.06$. At a higher density $\rho_{\text{pos}}$, there is a tetratic-solid transition which is also of Kosterlitz-Thouless type. The limiting forms, which indicates that short-range order will be detectable even in very small systems.

The phenomenological resemblance of the phase behavior to equilibrium systems appears to call for a thermodynamic description, by an extension of equilibrium statistical mechanics to non-equilibrium steady states. However, there are also important deviations from equilibrium behavior. In the moderate-amplitude regime, the evaporation density of the threefold cluster is larger than the density of the fluid coexisting with it. This dilution effect implies that pressure is non-monotonic as a function of $\rho$. In equilibrium this could only be possible in metastable states stabilized by nucleation barriers. Otherwise, the free energy would be non-convex, and thus inconsistent with the postulates of equilibrium statistical mechanics. As the evaporation density was compared instead of nucleation density, a nucleation barrier can be excluded. Therefore the dilution must be a non-equilibrium effect caused by the persistent energy flows and dissipation. A consequence of the anomalous dilution is a peculiar topology of the phase diagram, which features an ordered square phase intervening between the liquid and the liquid-threefold coexistence. In this region of the phase diagram, an effective free energy, or analogs of pressure and temperature that define phase equilibria, may not exist. Furthermore, it is an interesting question whether the fluid-threefold coexistence satisfies the lever rule for phase coexistence in equilibrium. The coexistence densities may be nontrivial functions of global density $\rho$ or of the size of the threefold cluster. Similar effects have been observed for active Brownian particles with hydrodynamic interactions \cite{note1}. We leave this point to a future study.

As a final non-equilibrium effect, traveling density waves and nontrivial particle currents associated with directed motion of stripe-shaped threefold clusters were found. The same mechanism is responsible for the faceting of the freestanding threefold cluster at low $A$. These freestanding clusters, however, do not exhibit directed motion and drift only diffusively. Future studies should clarify the underlying symmetry breaking in the microscopic dynamics and characterize the resulting energy and particle currents. The absence of rotational degrees of freedom makes this model a good starting point for tracking energy flows.

**ACKNOWLEDGMENTS**

TS was supported by the Deutsche Forschungsgemeinschaft as part of the Forschergruppe GPSRS under grant ME1361/13-2. We are grateful to Marcus Bannerman for providing the DynamO MD simulation program and for advice concerning the adjustment of the code to our needs, and to Uwe Täuber for helpful comments on an earlier version of the article. We thank Klaus Mecke for useful discussions and continued support.

\[\text{[1]}\] J. S. Olafsen and J. S. Urbach, Phys. Rev. Lett. 81, 4369 (1998).
\[\text{[2]}\] X. Nie, E. Ben-Naim, and S. Y. Chen, Europhys. Lett. 51, 679 (2000).
\[\text{[3]}\] J. S. Olafsen and J. S. Urbach, Phys. Rev. Lett. 95, 090602 (2005).
\[\text{[4]}\] E. Khain and I. S. Aranson, Phys. Rev. E 84, 031308 (2011).
\[\text{[5]}\] A. Prevost, P. Melby, D. A. Egolf, and J. S. Urbach, Phys. Rev. E 70, 056301 (2004).
\[\text{[6]}\] M. E. Lobkovsky, F. Vega Reyes, and J. S. Urbach, Eur. Phys. J. Special Topics 179, 113 (2009).
\[\text{[7]}\] W. Losert, D. G. W. Cooper, J. Delour, A. Kudrolli, and J. F. Gollub, Chaos 9, 682 (1999).
\[\text{[8]}\] J. S. Olafsen and J. S. Urbach, Phys. Rev. E 60, R2468 (1999).
\[\text{[9]}\] A. Kawarada and H. Hayakawa, J. Phys. Soc. Jpn. 73, 2037 (2004).
\[\text{[10]}\] N. Rivas, S. Ponce, B. Gallet, D. Risso, R. Soto, P. Cordero, and N. Mujica, Phys. Rev. Lett. 106, 088001 (2011).
\[\text{[11]}\] N. Rivas, P. Cordero, D. Risso, and R. Soto, New J. Phys. 13, 055018 (2011).
\[\text{[12]}\] N. Rivas, P. Cordero, D. Risso, and R. Soto, Granular Matter 14, 157 (2012).
\[\text{[13]}\] A. Prados and E. Trizac, Phys. Rev. Lett. 112, 198001 (2014).
\[\text{[14]}\] E. Trizac and A. Prados, Phys. Rev. E 90, 012204 (2014).
\[\text{[15]}\] J. J. Brey, M. I. García de Soria, P. Maynar, and V. Buzón, Phys. Rev. E 90, 032207 (2014).
\[\text{[16]}\] R. Brito, D. Risso, and R. Soto, Phys. Rev. E 87, 022209 (2013).
[17] P. Melby, F. Vega Reyes, A. Prevost, R. Robertson, P. Kumar, D. A. Egolf, and J. S. Urbach, J. Phys.: Condens. Matter 17, S2689 (2005).
[18] S. J. Moon, J. B. Swift, and H. L. Swinney, Phys. Rev. E 69, 031301 (2004).
[19] M. Schmidt and H. Löwen, Phys. Rev. E 55, 7228 (1997).
[20] P. M. Reis, R. A. Ingale, and M. D. Shattuck, Phys. Rev. Lett. 96, 258001 (2006).
[21] M. G. Clerc, P. Cordero, J. Dunstan, K. Huff, N. Mujica, D. Risso, and G. Varas, Nat. Phys. 4, 249 (2008).
[22] F. Vega Reyes and J. S. Urbach, Phys. Rev. E 78, 051301 (2008).
[23] M. Guzmán and R. Soto, Phys. Rev. E 97, 012907 (2018).
[24] G. Castillo, N. Mujica, and R. Soto, Phys. Rev. Lett. 109, 095701 (2012).
[25] G. Castillo, N. Mujica, and R. Soto, Phys. Rev. E 91, 012141 (2015).
[26] M. N. Bannerman, R. Sargant, and L. Lue, J. Comput. Chem. 32, 3329 (2011).
[27] N. V. Brilliantov and T. Pöschel, Kinetic theory of granular gases (Oxford University Press, 2010).
[28] W. Mickel, S. C. Kapfer, G. E. Schröder-Turk, and K. Mecke, J. Chem. Phys. 138, 044501 (2013).
[29] To be published.
[30] B. I. Halperin and D. R. Nelson, Phys. Rev. Lett. 41, 121 (1978).
[31] A. P. Young, Phys. Rev. B 19, 1855 (1979).
[32] S. T. Bramwell and P. C. W. Holdsworth, J. Phys.: Condens. Matter 5, L53 (1993).
[33] J.-P. Hansen and I. R. McDonald, Theory of Simple Liquids edited by Oxford (Academic Press, 1990).
[34] J. M. Kosterlitz, J. Phys. C: Solid State Physics 7, 1046 (1974).
[35] E. P. Bernard and W. Krauth, Phys. Rev. Lett. 107, 155704 (2011).
[36] S. C. Kapfer and W. Krauth, Phys. Rev. Lett. 114, 035702 (2015).
[37] V. L. Berezinskii, Sov. Phys. JETP 32, 493 (1971).
[38] D. R. Nelson and B. I. Halperin, Phys. Rev. B 19, 2457 (1979).
[39] J. Blaschke, M. Maurer, K. Menon, A. Zöttl, and H. Stark, Soft Matter 12, 9821 (2016).