Turbulent route to two-dimensional soft crystals

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We investigate the effects of a two-dimensional, incompressible, turbulent flow on mono-disperse soft granular particles and show the emergence of a crystalline phase due to the interplay of Stokesian drag (measured through the Stokes number) and short-range inter-particle interactions. We quantify this phase through the bond order parameter and local density fluctuations and find a sharp transition between the crystalline and non-crystalline phase as a function of the Stokes number. Furthermore, the nature of preferential concentration, as characterised by the radial distribution function and the correlation dimension $D_2$, is significantly different from that of particle-laden flows in the absence of repulsive potentials.

The self-assembly of particles in a flow [1-2], because of its ubiquity, is amongst the most studied problem in the areas of turbulent transport, soft matter, granular systems and nonequilibrium statistical mechanics. In quiescent form, most dilute assemblies are liquids, which when densified, can take a crystalline or amorphous structure depending upon the dispersity of the constituents [3-6]. In recent days, extensive studies of the rheology of such suspensions have happened [7-8], motivated by diverse applications. In typical experiments and computer simulations, the role of a carrier flow in dispersing the particulate matter is trivial: indeed if there is an underlying fluid medium, they are typically simple shearing [9-10]. In a variety of natural and industrial processes, however, particles are dispersed in flows with non-trivial spatio-temporal correlations which are chaotic, and in extreme cases (such as a marine system) even turbulent [11-13].

This specific question of the structural properties of particulate suspension, where the underlying flow is turbulent, has surprisingly been not investigated despite significant progress in the last two decades in the area of turbulent transport of finite-sized, heavy, inertial (colloidal) particles. In this paper, we report the emergence of macroscopic particulate structures with crystalline (hexagonal) motifs even in the presence of strong mixing because of the carrier turbulent flow.

For suspended particles with a finite diameter and density, inertial effects and dissipative dynamics become important, leading to the particles detaching from the underlying flow to form strong inhomogeneities in their spatial distribution (see Fig. 1). This phenomenon, known as preferential concentration, has been extensively studied [14-19] and remains critical to our explanations of problems such as rain initiation in warm clouds. Although more recent studies have addressed the issue of problems such as gravity [20-22] and turbophoresis [23-25] on preferential concentration, inter-particle interactions have largely been ignored except for studies on coalescences [26-27] and on Vicsek ordering [28]. The only example that we are aware of, which links ideas of soft matter and turbulent transport is the use of repulsive, elastic, hard sphere inter-particle interactions, which, combined with a dissipative dynamics, lead to stickiness and aggregation [29]. However, for most physical systems, the elastic limit is an idealised one: The most obvious particulate exchanges, such as those mediated through a soft potential [30] has been ignored so far.

In this work, we therefore address two important and related issues, namely what is the effect of soft particle interactions on clustering of particles in a turbulent flow and can such realistic interactions, contrary to naïve expectations, lead to the growth of stable crystalline structures in an ensemble of particles interacting with each other as well as an ambient turbulent fluid.

We consider an assembly of $N_p$ particles seeded in a two-dimensional, statistically stationary, turbulent velocity field $\mathbf{u}$. Since we consider particles smaller than the relevant length scales of the flow, the dynamics of the $i$-th particle (characterised by its Stokes or particle response time $\tau_p$) defined through it position $\mathbf{x}_i$ and velocity $\mathbf{v}_i$ is given by the linear Stokes drag model along with the inter-particle interaction potential $V(r_{ij})$:

$$
\frac{d\mathbf{x}_i}{dt} = \mathbf{v}_i; \quad \frac{d\mathbf{v}_i}{dt} = \frac{\mathbf{v}_i - \mathbf{u}(\mathbf{x}_i, t)}{\tau_p} - \sum_{j=1}^{N_p} \nabla V(r_{ij});
$$

(1)

where the interacting short-ranged repulsive potential [30-31], commonly used for modelling emulsions and other soft granular suspensions, is given by:

$$
V(r_{ij}) = \begin{cases} 
\frac{1}{2}(1 - r_{ij}/\sigma_{ij})^2 & \text{for } r_{ij} < \sigma_{ij}, \\
0 & \text{for } r_{ij} \geq \sigma_{ij};
\end{cases}
$$

(2)

where, $r_{ij}$ is the inter-particle separation, $\sigma_{ij}$ is the sum of the radii of particles $i$ and $j$, $\epsilon$ sets the energy scale for particle interactions. Thus, such an interaction takes into account the energy cost of deformation, only when two particles are in contact. For our work, we mostly consider a mono-disperse assembly, where all particles have the same diameter $\sigma$.

The advecting turbulent velocity $\mathbf{u}$ is obtained (by using a standard pseudo-spectral method) as a solution of
FIG. 1. Representative snapshots of interacting (red) and non-interacting (blue) particles with Stokes numbers (a) \( St = 0 \) (b) \( St = 0.75 \) and (c) \( St = 2.50 \), superimposed on the vorticity field of the carrier turbulent flow. Here \( \phi = 0.1\% \).

The two-dimensional, incompressible Navier-Stokes equation, on a \( 2\pi \) periodic grid, and a deterministic forcing to maintain a non-equilibrium steady state. The flow is conveniently described by its characteristic length \( l_\nu = \langle \sqrt{\Omega/P} \rangle \) and time \( \tau_f = \langle 1/\Omega \rangle \) scales, where \( \Omega = \int k^2 E(k) dk \) is the enstrophy and \( P = \int k^4 E(k) \) the palinstrophy \[32\]. This allows us to naturally define the non-dimensional Stokes number \( St = \tau_p/\tau_f \) and ensure that both the grid spacing and particle diameter are much smaller than \( l_\nu \). It is convenient to vary the Stokes number by using different values of \( \tau_p \); the elasticity or softness of the interactions are tuned by varying \( \sigma \), keeping the energy scale fixed at \( \epsilon = 1 \).

The effect of such inter-particle interactions is striking. In Fig. 1 we show representative snapshots of particle positions, superimposed on the background vorticity field, for both interacting (red) and non-interacting (blue) particles. In the absence of inertia (\( St = 0 \)), unsurprisingly, the difference between the two ensembles is minute (see panel (a) on Fig. 1). However, for finite values of \( St \) (panels (b) and (c)), the particle distributions are strongly influenced by their interactions. In particular, the nature of small-scale clustering is significantly altered and, especially for \( St > 1 \) (panel (c)), interacting particles appear to be more homogeneously distributed than the ones which are non-interacting.

Inhomogeneities in the particle distribution are conveniently characterized by the correlation dimension \( D_2 \), defined through the probability of having two particles within a distance \( r \), namely \( P^{<}_2(r) \sim r^{D_2} \), or equivalently through the small-scale behavior of the radial distribution function \( g(r) \sim r^{D_2-2} \). Figure 2 (top) shows \( D_2 \) as a function of \( St \) (including the non-interacting case), for various values of the packing fraction \( \phi = N_p \sigma^2/(16\pi) \). We fix \( \sigma = 5.0 \times 10^{-4} \), and choose \( N_p = 1 \times 10^5 \), \( 1.5 \times 10^5 \) and \( 2.0 \times 10^5 \), to obtain \( \phi = 0.05\%, 0.075\% \) and \( 0.10\% \), respectively. For a given packing fraction and \( St \ll 1 \), the value of \( D_2 \), within error-bars, are indistinguishable from the non-interacting particles. This is because at such small values of \( St \), there is hardly any small-scale clustering and therefore such dilute suspensions are only weakly affected by the short-range particulate interactions. However, as particles cluster, i.e. for \( St = \mathcal{O}(1) \), soft granular repulsions dominate and, unlike

![FIG. 2. Correlation dimension \( D_2 \) vs \( St \) for non-interacting (blue triangles) and interacting particles for ensembles for \( \phi \), where \( N_p \) varies (see legend). Inset: Correlation dimension \( D_2 \) vs \( St \) for non-interacting (blue triangles) and interacting particles \( \phi \) where \( N_p \) is fixed. (The error-bars are smaller than the symbols size.)](image-url)
the non-interacting ensemble, the interacting particles spread more — as a result of the competing interactions of vortical-ejection due to inertia and the short-range inter-grain repulsive energy cost — and with a larger value of $D_2$. This effect also leads to a slight shifting to the left of the value of $St$ where $D_2$ attains its minimum. This effect is of course accentuated with increasing packing fractions. For Stokes numbers a bit larger than 1, although the centrifugal vortical-ejection weakens, particles still tend to cluster in straining zones. However, the strength of the short-range repulsive forces ensure that interacting particles spread out more and sample the flow homogeneously, overcoming the bias due to inertia (Fig. 1(c)). We see evidence of this in our measurements, which show that for $St > 1$ the correlation dimension $D_2$ asymptotes to the physical dimension 2 much faster for the interacting than for the non-interacting case.

In the absence of the fluid, fixing size or number doesn’t matter — the physics is the same. So, it is the influence of the flow that makes the difference, in this case. It is important to examine the role of the softness or elasticity of the interacting potential which depends on the particle diameters. This is best demonstrated by keeping $N_p$ fixed, but using different values of $\sigma$. We now choose $N_p = 1 \times 10^5$ and $\sigma = 5.0 \times 10^{-4}$, $10.0 \times 10^{-4}$ and $\sigma = 15.0 \times 10^{-4}$. As the diameter increases, particles become softer and more inelastic; Hence when they collide, they tend to stick and bounce off less from one other. Thus we would expect that for softer particles, $D_2$ for the interacting and non-interacting cases should be the same especially for $St = O(1)$. However this effect is slightly off-set by the fact that an increase in particle diameters, and hence their softness, leads to a larger collision frequency and more chances of particles undergoing enough collisions to separate. The inset of Fig. 2 shows a plot of $D_2$ for different particle diameters. The observed behavior seems to be consistent with this conjecture: Increasing $\sigma$ makes particles more and more adhesive and hence $D_2$ gets closer to the values obtained for the non-interacting case.

Before we turn to the structural aspects of this particulate system, it is important to appreciate this rather curious interplay of particle diameters, the effect of packing and Stokes numbers — suggestive of a complicated, non-monotonic phenomenon — purely from the point of view of a turbulent transport problem. Indeed to bring out the salient features of this effect, we chose to make our particle diameters independent of $St$. We have however checked that if we indeed change the $\sigma \propto \sqrt{St}$, the resulting inhomogeneity in particle distribution is entirely consistent with the conclusions drawn from Fig. 2.

Inter-particle interactions in particle-laden turbulent flows clearly have an effect on the degree and nature of preferential concentration. But are such particle interactions strong enough to overcome turbulent mixing and nucleate crystalline structure? To answer this question — and provide compelling evidence — it is essential to work with a much larger packing fraction and particle diameter, as is common in studies of granular systems [33, 34]. (We however ensure that the diameters are still much smaller than the fluid characteristic length scale $l_v$ for our model to be valid.) We therefore choose $\sigma = 5 \times 10^{-3}$, and different particle numbers, namely, $N_p = 2 \times 10^5$ ($\phi = 10\%$) and $N_p = 10^6$ ($\phi = 50\%$).

A useful indicator of how densely such particles are packed, due to inertia and interactions, is to look at the local packing fraction $\phi_{\text{local}} = N_\Delta \pi \sigma^2/(4 \Delta^2)$, where $N_\Delta$ is the number of particles in a small square of side $\Delta = 4 \delta x$, where $\delta x$ is the width of our Eulerian grid. In Fig. 3(a) we show a representative pseudo-color snapshot of the local packing fraction for particles with $St = 1$ ($\phi = 50\%$). As we would expect, in a given snapshot, there are regions which are extremely dense and the local packing fraction far exceeds its average $\phi$, with some regions hyper-packed due to the softness of the particles. We now examine the structure of these densely packed regions by
using the standard approach of Voronoi tessellation. In Fig. 3(c) we show the Voronoi construction, corresponding to a zoomed in region of panel (a); furthermore we color, on a greyscale, each cell by the coordination number $z$ (values shown in the adjacent colorbar) of the particle. We find, surprisingly, that these soft particles do form hexagonal lattices, as suggested by the predominance of $z = 6$ (light grey) and the cells of our Voronoi tessellation, with almost perfect crystalline order. For extremely small values of the Stokes number, $St$, we have a re-entrant melting scenario \[36\].

To quantify this degree of crystallinity, we use the standard measure of the bond order parameter $\psi_6$ \[35\] for a given particle:

$$\psi_6(r_i) = \frac{1}{N_b} \sum_{m=1}^{N_b} \exp[6 \cdot \theta_{mi}], \quad (3)$$

$N_b$ is the number of nearest neighbors of the $i$th particle, and $\theta_{mi}$ is the angle between the $x$-axis and the bond joining the $i$th particle with the $m$th particle. Before we turn to the full statistics of the bond order parameter, it useful to first look at a coarse-grained measure of this quantity. In analogy to our definition of a local packing fraction, we define a local bond order parameter $\psi_6^{\text{local}} = \langle 1/N_b \rangle \sum_{i=1}^{N_b} \psi_6(r_i)$. The map for $\psi_6^{\text{local}}$, corresponding to the packing shown in Fig. 3(a), is displayed in Fig. 3(b). Such a coarse-grained description shows macroscopically large regions with a very high value of $\psi_6^{\text{local}}$ consistent with the visual suggestion of crystallinity in Fig. 3(c). Also, note that whenever the soft particles are hyper-packed, the crystallinity is lost, and we have a re-entrant melting scenario \[36\].

The snapshots of Fig. 3 naturally lead us to examine the behaviour of $\psi_6$ as a function of $St$ for a reasonably high packing fraction ($\phi \sim 10\%$). Given the non-equilibrium, spatio-temporal variation of the advecting turbulent flow, it is of course natural that all the particles would not arrange themselves in a hexagonal lattice. We therefore look at the mode $\langle \psi_6 \rangle$ of the values of $\psi_6$ for all particles and over time. In the upper panel of Fig. 4 we plot $\langle \psi_6 \rangle$ (blue filled circles) as a function of $St$ and see a remarkable behaviour. For extremely small or large values of the Stokes number, $\langle \psi_6 \rangle = 0$ whereas for values of $St$ around 1, where, as seen in Fig. 2 particles show significant preferential concentration, $\langle \psi_6 \rangle = 1$. This behaviour is remarkable as it shows a sharp transition between a crystalline and non-crystalline phase (as a function of the Stokes number). Of course such a characterisation makes sense only if there is a macroscopically large fraction of the total particles which show $\langle \psi_6 \rangle$ as reported in Fig. 4 (Top). We thus calculate the fraction of particles (along with their errorbars calculated over time) having $\psi_6 = \langle \psi_6 \rangle$ (magenta square symbols). We immediately see that in the crystalline phase nearly 20% of all particles arrange themselves in perfect hexagonal order. If we use definition that $\psi_6 = [0.80 \ 1.0]$ corresponds to local crystallinity, the fraction of particles in such patches corresponds to 50%, indicating that indeed there is a dynamical structural transition with Stokes number. These two curves, together, give quantitative evidence for the emergence of a macroscopic crystalline order in an ensemble of particles in a turbulent flow. In inset of the same figure, we also plot the mean bond order parameter $\langle \psi_6 \rangle$ as a function of the Stokes number. The non-monotonic behaviour of this plot is consistent with that seen for $\langle \psi_6 \rangle$. (Of course the average value does not switch between 0 and 1 because of local fluctuations in the particle arrangement.)

Finally, we examine if there is evidence for a similar sharp transition, as the overall particle density is increased. In the lower panel of Fig. 4 for $St = 1$ — the regime where crystalline structures are observed, as shown in the top panel — we find good evidence for onset of crystallinity with increasing $\phi$, by measuring how $\langle \psi_6 \rangle$ varies. Indeed, for low packing fractions, this measure shows that local structures are more liquid-like before saturating, within error-bars, at values consistent with the presence of macroscopic hexagonal structures, as discussed before.

In conclusion, we have shown that two-dimensional particle-laden turbulent flow, lead to crystalline self-assembly, because of the complementary effects of drag-induced preferential concentration and inter-particle in-
The central role played by particle inertia is apparent in the sharp transition between crystalline and non-crystalline aggregates as a function of the Stokes number. Our work also shows that elasticity of the particles lead to a modification in the nature of preferential concentration of heavy inertial particles which lie at the heart of several natural and industrial processes.

It is also important to keep in mind that, given that there have hardly been any studies of particulate structures in a turbulent flow, our work focuses on the most simple and general framework as commonly used in turbulent transport problems. The most important simplification that we have used is to ignore the feedback of the particles on the flow, as well as lubrication forces or the effect of porosity in the packed structures. It has been shown in an earlier work that a one-way coupled model for Stokesian particles is a valid assumption in turbulent flows. Furthermore, it was shown in [29], while studying the problem of elastic collisions in particle-laden turbulent flows, that at least in the small Stokes limit, the effect of short-range lubrication was merely the renormalisation of the effective relaxation time. However, it should be left for future work to actually examine in detail the role of lubrication and porosity in stabilising such crystalline structures.

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