Oscillatory driven colloidal binary mixtures: axial segregation versus laning

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Using Brownian dynamics computer simulations we show that binary mixtures of colloids driven in opposite directions by an oscillating external field exhibit axial segregation in sheets perpendicular to the drive direction. The segregation effect is stable only in a finite window of oscillation frequencies and driving strengths and is taken over by lane formation in direction of the driving field if the driving force is increased. In the crossover regime, bands tilted relative to the drive direction are observed. Possible experiments to verify the axial segregation are discussed.

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Phase transitions in driven systems are qualitatively different from their equilibrium counterparts and reveal a wealth of novel instabilities and pattern formations induced by nonequilibrium conditions \([1]\). In particular, it is very intriguing to resolve and follow nonequilibrium dynamics on the particle level which is possible for granular grains \([2,3]\), mesoscopic colloidal suspensions \([4]\) or dusty plasmas driven by an external field \([5]\).

In the colloidal context, recent investigations have focused on binary mixtures which are driven by a constant but species-dependent force. The latter can be realized by gravity \([6,7]\) or by an electric field \([8]\). If the drive is strong enough, lanes of particles driven alike are formed in direction of the driving field \([9]\).

In this paper we simulate a model for a colloidal mixture with an time-dependent oscillatory drive \([28]\). We show that this induces an axial segregation, i.e. an ordering in sheets normal to the driving field (rather than parallel as for laning). While such an axial segregation of two particle species is observed in shaken or vibrated granular systems \([10,11,12,13]\), it has not yet been described for colloidal suspensions whose dynamics is dominated by overdamped Brownian motion. This makes the colloidal dynamics different from that of granular systems where inelastic collisions play an important role. At fixed density, we map out the whole non-equilibrium steady-state diagram over a large range of frequencies and driving amplitudes and find a rich phase diagram topology. By increasing the magnitude of the driving force or decreasing the oscillation frequency, the system gets back either to a disordered state or to lane formation with an intermediate regime where mixing of segregation and laning as well as tilted lanes occur. Decreasing the magnitude of the driving or increasing the oscillation frequency, on the other hand, always yields a disordered steady state. We then study the relaxational dynamics into the steady states and show that first laning occurs and if at all axial segregation occurs on a larger time scale. Finally we discuss the anisotropy of the mean-square-displacements in the different steady states as dynamical consequences of their different structure.

In order to simulate the dynamics of a colloidal suspension we utilise a two dimensional (2D) Brownian dynamics (BD) simulation with neglected hydrodynamic interactions between the particles. In particular, we consider an equimolar binary mixture of \(N = 2864\) hard disks of diameter \(\sigma\). In Brownian dynamics, the time evolution of the individual colloids is governed by overdamped Langevin equation with thermal noise \([14]\). The particles of sort \(A\) and \(B\) are subjected to an oscillatory inversely phased force \(f^A(t) = -f^B(t) = f_0 \sin(\omega t) e_x\), where \(\omega\) is the driving frequency and \(f_0\) the driving strength. A single particle will follow a noise-averaged trajectory which is sinusoidal in time with an amplitude \(f_0/\xi \omega\).

We simulate a quadratic system of size \(L/\sigma = 75\) with periodic boundary conditions at a total area fraction \(\phi = N \pi \sigma^2/(4L^2) = 0.4\). As a measure for the driving strength, we define the Peclet number \(Pe = \tau_D/\tau_d\) as the ratio between the time \(\tau_D = \sigma^2/(4D)\) it takes a colloid to diffuse its own radius and the time \(\tau_d = \sigma/(2v_d)\) it takes to drift the same distance under the action of a constant external force \(f_0\). Here \(D = k_B T/\xi\) denotes the short-time diffusion constant where \(k_B T\) is the thermal energy, \(\xi\) the friction constant and \(v_d = f_0/\xi\) is the maximal drift velocity.

The initial configuration of the BD simulation is an equilibrated fully mixed configuration as realized in the absence of the drive. At time \(t = 0\), the oscillatory force is turned on instantaneously and the evolution of the system is followed by using a finite time-step algorithm \([15]\) up to about \(10^5\) oscillation periods. Depending on the Peclet number \(Pe\) and the reduced frequency \(\omega \tau_D\), the system ends up in different steady states.

The nonequilibrium steady-state phase diagram is shown in Fig.\([1]\) spanning two decades in both driving frequency \(\omega \tau_D\) and strength \(Pe\). Coresponding movies showing the development from the initial to the steady state are available in \([29]\). Let us discuss the topology of the phase diagram step by step. For very low driving strengths, the external drive is not strong enough to generate order in the completely mixed equilibrium state leading to a disordered steady-state which is depicted
in a typical snapshot in Fig. 1a. A disordered state is rendered as crosses in the full phase diagram shown in Fig. 1e such that the region of low Peclet numbers involves a disordered steady state. As visible in Fig. 1a, the disordered state can possess an intrinsic finite correlation length as set by colliding clusters which is larger than the interparticle distance but these structures do not span the whole simulation box. For very high frequencies, on the other hand, a free particle would just perform oscillations with a very small amplitude due to the drive. This amplitude $v_d / \omega$ is indicated as an inset in the corresponding simulation snapshots in Fig. 1a-d. At high frequencies, an ensemble of many particles, is just rattling over a small distance and is thus not perturbing much the disordered equilibrium state. Conversely, at very low frequencies, lane formation similar to that observed earlier for a static or slowly oscillating drive occurs where the direction of the lanes coincides with the drive direction. A corresponding simulation snapshot is given in Fig. 1d. In a region of finite frequencies and Peclet numbers, bounded by disordered and laned steady states, axial segregation shows up, where the stripes are oriented perpendicular to the drive direction, see the corresponding snapshot in Fig. 1b. Here the particles driven alike perform collectively oscillations as induced by the external drive but collide periodically with an opposing band of opposite particles. In Fig. 1b, a colliding interface is seen in the middle of the snapshot while voids in the two other interface indicate that the bands had just been driven away from this position. We call this stripe formation perpendicular to the driving field axial segregation. Finally, in an intermediate region between axial segregation and laned, some steady states are observed with tilted bands, see e.g. the snapshot in Fig. 1c, where a general tilt angle $\theta$ relative to the drive direction is observed. However, as is discussed in detail below, the major part of the steady states are still either segregated or laned.

There are two reentrant effects of the disordered phase in the phase diagram shown in Fig. 1e: first by increasing the Peclet number at fixed frequency $\omega_{TD} \approx 1$, the steady state transforms from the disordered one into the axially segregated one and back to the disordered one. Second, for increasing frequency at fixed Peclet number $Pe < 5 \approx 10$, the disordered phase is taken over by the axially segregated phase and then appears again.

A simple quantitative criterion for the disordered-to-segregated transition at high frequencies can be derived by the intuitive argument that the oscillating amplitude $v_d / \omega$ must exceed the mean interparticle distance $\sigma(\sqrt{\Theta_{max}} / \sigma - 1)$ in order to induce a noticeable segregation effect. The analytical estimate of this phase boundary line is shown as a solid line in Fig. 1e and agrees well with the simulation data at intermediate and high Peclet numbers.

We now turn to the dynamics of how the different steady states are reached after turning on the driving field. For this purpose we stroboscopically determine the location of the colloids after each drive period $T$ (at a time where all external forces vanish) and calculate the partial static structure factor $S(k, nT) = ...$
defined as the final segregated state. This clearly shows that the for-
after this induction time that the system goes into the
is persistent over more than a decade in time. It is only
and four different end states two two of which are segre-
gated end-state and one is laned and tilted. Strikingly ,
structure (\(\theta\)) and define \(\omega\) as well as the angular frequency
fixed \(c\) and \(\theta\) with \(\alpha\) ∈ \(\{x, y\}\) and \(k_{\text{max}} = 2\pi/\sigma\). Here \(r_i\) with \(i \in \{1, \ldots, N/2\}\) denote the positions of all partic-
les of the same species. From the structure factor we get
the typical length scale of the system via a first moment defined as
\[
\frac{2/N}{l_{\text{t}}(nT)} = \frac{\int_{k_{\text{max}}}^{k_{\text{max}}} |k_{\alpha}| S(e_{\alpha} k, nT) dk_{\alpha}}{\int_{-k_{\text{max}}}^{k_{\text{max}}} S(e_{\alpha} k, nT) dk_{\alpha}} \quad (1)
\]
To obtain the orientation of a typical structure in the system with respect to the direction of the oscillatory force we calculate the tensor
\[
S = \int_{k_{\text{max}}}^{k_{\text{max}}} S(k, nT) [(kk)I - k \otimes k] dk
\quad (2)
\]
(where \(I\) denotes the unit tensor and \(\otimes\) the dyadic prod-
uct) and define \(\theta\) as the angle between \(e_x\) and the eigen-
vector \(e_1\) corresponding to the biggest eigenvalue of \(S\).
The angle \(\theta\) serves as an "order parameter" of the ob-
served structures: in fact, \(\theta = 0\) points to a state of lanes
while \(\theta = \pi/2\) corresponds to axial segregated structures
and \(0 < \theta < \pi/2\) indicates a tilted state.

The time evolution of the two length scales \(l_x\) and \(l_y\)
well as the angle \(\theta\) is shown in Fig 2 at fixed driv-
ing frequency \(\omega = 4\) for three different Peclet numbers
and four different end states two two of which are segre-
gated end-state and one is laned and tilted. Strikingly ,
the system first develops structures along the drive which
is persistent over more than a decade in time. It is only
after this induction time that the system goes into the
final segregated state. This clearly shows that the for-
mation of the axially segregated steady state is a highly
collective phenomenon which cannot by accounted for by
a local instability analysis. Laning, on the other hand,
can be viewed as a local instability \(16, 17\).

Let us now discuss the tilted state in some more de-
tail: in the range \(30 < Pe < 80\) some steady states pos-
sess a non-trivial winding around our "toroidal" system
(which is dictated by the periodic boundary conditions
in the two dimensional system), i.e. \(\theta = \arctan(n/m)\)
with natural numbers \(n, m\) and more precisely we observe
\(n/m = 1/2, 1/1, 2/1\). We remark that similar steady
states were also found in constant driven diffusive lattices
gases \(1, 18\) and in binary mixtures which are driven by
nonparallel external forces \(19\). We observe that with in-
creasing \(Pe\) the probability to end in a tilted steady state
with \(\theta = \pi/4\) increases and gets nearly equal to that of
an axially segregated \((\theta = \pi/2)\) state for \(Pe = 60\). In the
range \(50 < Pe < 60\) there exists even a finite probability
for a third steady state \(\theta = \arctan(2)\). On average the
tilted state is reached after \(10^2 - 10^3\) periods and was
always stable during further \(10^5\) periods. We checked
that doubling the system size yields again a tilted state
with same angle \(\theta\). Though this clearly shows that tilting
is persistent, we cannot exclude at this stage that the tilting
is an artifical finite-size effect stemming from the
toroidal boundary conditions. However, one can spec-
ulate \(18\) that the lifetime of the tilted states diverges
with system size. If so, this would be an example for
multistability in a finite region of the \(Pe - \omega T_D\) space as
a consequence of nonequilibrium dynamics \(20\).

We finally monitor the mean-square-displacement as a
function of time \(t = nT\) in the region of multistability

![FIG. 2: Time evolution of a typical length scale parallel (a) and perpendicular (b) to the direction of the oscillatory force \(l_x/\sigma\) and \(l_y/\sigma\), as well as of the orientation \(\theta\) of a typical structure (c). In the BD simulations the angular frequency is fixed \(\omega T_D = 4\) and the Peclet numbers are \(Pe = 20, 50, 120\). For \(Pe = 50\), both a tilted and segregated phase are shown.](chart1.png)

![FIG. 3: Distribution of the orientation \(\theta\) of the steady state structures starting from a fully mixed configuration after a simulation time of \(10^4 T\). For every Peclet number 200 independent simulations have been performed. The angular frequency is fixed to \(\omega T_D = 4\).](chart2.png)
we notice that for θ

rent along the tilted interface in driven lattics gases \[18\],

in xy MSD does not vanish ∆

collisions which increase the particle mobility. Moreover,

able during the first 10

square displacement of the two systems is indistinguish-

axially segregated state with the tilted state, the mean

pendicular to the structures formed. In comparing the

if the mean-square-displacement is taken for motion per-

ular to the drive for appropriate frequencies and driv-

θ

constant), while for

θ

and define the corresponding displacement correlations:

\[ \Delta_{\alpha\beta}(t) = 1/N \sum_{i=1}^{N} (\Delta r_i(t) \cdot e_{\alpha})(\Delta r_i(t) \cdot e_{\beta}) \] (3)

where \( \Delta r_i(t) = r_i(t) - r_i(0) \) and \( \alpha, \beta \in \{x, y\} \). More precisely we consider systems which end up in one of the three steady states with \( \theta = 0, \pi/4, \pi/2 \). The two cases \( \pi/4, \pi/2 \) are calculated at the same Peclet number \( Pe = 50 \). Results for \( \Delta_{\alpha\beta}(t) \) are presented in Fig.4 on a double-logarithmic scale. We conclude that for small times the motion is diffusive while for long times it is either diffusive or bounded. A quasi-bounded motion arises if the mean-square-displacement is taken for motion perpendicular to the structures formed. In comparing the axially segregated state with the tilted state, the mean square displacement of the two systems is indistinguishable during the first \( 10^2 \) periods, but enhanced compared to a free colloid due to enormous number of interparticle collisions which increase the particle mobility. Moreover, \( \Delta_{xx} > \Delta_{yy} \) for small times due to initial local laning. For \( n > 10^2 \) and \( \theta = \pi/2 \), particles can unhindered diffuse in y direction and are trapped in x direction (\( \Delta_{xx} \) nearly constant), while for \( \theta = \pi/4 \) the particles diffuse equally in x and y direction. Comparable to a finite mass current along the tilted interface in driven lattices \[18\], we notice that for \( \theta = \pi/4 \) the offdiagonal part of the MSD does not vanish \( \Delta_{xy} \neq 0 \) which is a consequence of a shear-flow-like displacement field. This is opposed to \( \theta = 0, \pi/2 \) where \( \Delta_{xy} \) vanishes.

In conclusion, we have shown that two species of oscillatory driven colloids segregate into stripes perpendicular to the drive for appropriate frequencies and driving strengths. We have performed additional simulations which show that axial segregation is stable in various situations: i) for softer interparticle interactions, ii) in three spatial dimensions where the stripes are two-dimensional sheets of finite width, iii) in slit-geometry confinement when the driving is perpendicular or parallel to the slit, iv) if hydrodynamic interactions mediated by a solvent flow are included in the simulation and the physical volume fraction is small. The disordered state can be reentrant for increasing the driving frequency and strength. For increasing strength there is a transition towards lane formation delineated by a mixed situation of laning and segregation where tilted stripes do also occur.

In principle, it is possible to verify the segregation effect in real-space experiments on driven binary colloidal mixtures. Experimental realisations are superparamagnetic colloids driven by a gradient in a magnetic field \[21\], oppositely charged colloidal mixtures exposed to an alternating electric field \[8\], colloids driven by gravity \[6, 21\] in a rotating cell or colloidal mixtures driven by dielectrophoretic force in a nonuniform AC electric field \[22, 23\]. In particular we think that opposite charged colloids in an AC electric field are a very promising realization to see axial segregation \[30\]. Since hydrodynamic interactions are strongly screened in this case \[24\], our model suitably generalized to three spatial dimensions and Yukawa pair interactions should be appropriate to describe this system.

It would be interesting to explore higher densities where freezing occurs \[19\] and to investigate the solvent flow field which might strike back to the pattern formation at intermediate physical volume fractions. Finally, it is challenging to describe axial segregation by a microscopic theory based on the Smoluchowski equation where either dynamical density functional theories \[14, 17\] or mode-coupling approaches \[25\] could be used. We leave this important study for the future.

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[28] Recent investigations with oscillatory drives have revealed quite rich non-equilibrium transitions including e.g. drive-induced ergodicity breaking, see e.g. [24-27].
[29] Movies of the simulation can be viewed at http://www2.thphy.uni-duesseldorf.de/~adam/.
[30] The jamming in bands perpendicular to the field discussed in Ref. [8], however, is different from segregation since a DC electric field was applied.