ORIGINS OF REDUCTION IN EFFICIENCY IN MICROFLUIDIC PARTICLE SEPARATION

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ABSTRACT
Microfluidic particle separation often gets degraded by defocusing of particles at high concentration due to hydrodynamic particle-particle interactions. We theoretically and experimentally studied the origins of these interactions; the reversing wake reflected from nearby channel walls is found to be the dominant interaction. In addition, we present a unique mechanism for the dynamic self-assembly of particles in finite-Reynolds number channel flow. Inertial lift forces and a parabolic flow field act together to stabilize interparticle spacings that otherwise would diverge to infinity due to viscous wakes.

KEYWORDS: Particle separation, Hydrodynamic interaction, Inertial microfluidics, Dynamic self-assembly

INTRODUCTION
Various label-free techniques have been developed to separate and sort particles in microfluidic systems [1]. For example, separation of different size particles using inertial focusing provides simple operation and unprecedented high-throughput [2, 3]. However, hydrodynamic particle-particle interactions induce defocusing of particles resulting in lower efficiency for separations (Fig. 1a, b). Here we present the mechanism of defocusing dynamics which involves viscous wakes in confined geometries. In addition, we show that these viscous wakes and inertial lift forces work together to yield self-assembled particle systems in finite-Reynolds-number channel flows. Here, Reynolds number is defined as $Re = \frac{\rho U_m H}{\mu}$, where $\rho$ is the density of the fluid, $U_m$ is the maximum flow speed, $H$ is the hydraulic diameter, and $\mu$ is the dynamic viscosity of the fluid. Inertially focused particles in confined high-speed microfluidic channel flows dynamically self-assemble into uniformly spaced “lattices” (Fig 1c). We hypothesized that the viscous wake acts as a repulsive interaction while inertial lift forces stabilize the particles at finite distance. Due to inertial focusing [3] particles flowing in rectangular channels typically occupy two transverse equilibrium positions, while square channels yield four due to channel symmetry. We used two-inlet coflow with particle-free fluid to achieve 1-D lattices to minimize the degrees of freedom, and give a simplified system to study particle wakes and particle-particle interactions.

RESULTS AND DISCUSSION
Hydrodynamic particle-particle interactions broadly refer to various interactions elicited by the disturbance a particle causes to a flow field and that affects another particle. Engineering approaches to control these interactions are difficult unless the dominant disturbances can be identified. One of the hydrodynamic interactions that is found to be most relevant in confined microchannel geometry is the reversing wake reflected off nearby channel walls [4]. Defocusing dynamics of inertially focused particles can help understand the effect of channel confinement. In expanding channels, particles slow down and move closer due to expanding streamlines. Inertial lift forces decrease monotonically with increasing channel width while viscous interactions between particles increase due to decreasing interparticle spacing (scaling like a mass dipole). Due to this effect inertially focused particles “defocus” or migrate away from the streamlines that the particles were originally following (Fig. 1b).

We used inertially-focused particle pairs to study this phenomenon in more detail. Figure 2a shows a particle pair entering an expanding channel experiencing defocusing. Defocusing is clearly not due to a collision. Defocusing can be explained by a broken balance between the inertial lift forces and viscous drag from the wake of the nearby particle; the lagging particle is always pushed towards the wall, and the leading particle is pushed away from the wall (Fig. 2b). Note that the defocusing is a deterministic behavior (Fig. 2a inset), suggesting the possibility of controlling the defocusing motion with control of particle concentration and channel geometry.

Figure 1: Concentration dependent defocusing of microparticles (a) Inertially focused particles at the expansion follow single streamlines (dotted lines). (b) At higher concentrations particles move across streamlines and are defocused due to hydrodynamic particle-particle interactions. (c) Particles are inertially focused and ordered to form lattices. Co-flow with particle-free fluid on one side allows creation of a 1-D lattice.
We next investigated the wakes around single particles in a confined channel. Reversing wakes accompanying rotating particles have been suggested as a unique and unexpected aspect of flow around a sphere in finite-Reynolds-number shear flow [5, 6]. However, recently it was theoretically shown that reversing streamlines and swapping trajectory particle motion do not necessarily require fluid inertia but can occur in Stokes flow in a confined channel geometry [4]. The reversing wake in Stokes flow thus arises from the reflection of the disturbance flow off the channel boundary and does not require inertia. Here we extend these results, shown in linear shear flows, to parabolic channel flows. We have simulated a flow around a sphere at zero-Reynolds number and finite-Reynolds number using previously described numerical methods [7]. Zero-Reynolds number simulations are done by setting fluid density equal to zero while keeping the flow rate at 100 µL/min. The simulation results at Re = 0 (Fig. 3a) confirm that recirculation occurs from channel confinement in rectangular channel flow (parabolic shear). The simulation results for finite-Reynolds number gave nearly identical reversing streamlines with slight asymmetry due to inertia (data not shown). We have also experimentally observed the existence of these reversing flows for the first time (Fig. 3b). In the frame of reference of a larger particle, tracer particles are observed to reverse direction. This behavior was also observed regardless of particle Reynolds number, in agreement with the simulation results and suggesting the reversing flow is predominantly due to the confinement effect. Importantly, this finding makes this effect relevant to all microfluidic particle separation systems.

Self-assembling systems, in general, require multiple interactions that include positive and negative feedback, which for particle systems are realized as attractive and repulsive forces. We studied dynamics of interactions between two particles in the context of the previously discussed viscous hydrodynamic wakes (Fig. 4). Captured movies were transformed to x-t graphs to visualize the dynamics of the particle-particle interaction. Initially, particles at the inlet move with different speeds because they are randomly distributed over a parabolic velocity profile. Due to differences in speed, a faster particle approaches a slower particle and forms a particle pair that moves downstream together. Particle pairs show various dynamics when the distance between the two particles is small enough for the particle-particle interaction to become significant (< ~100 µm). In Figure 4a, four particle pairs and a single particle are shown. Surprisingly, particle pairs undergo a variety of behaviors including oscillatory motion in the x direction before they settle to an organized state.

Dynamics of pair-wise particle interactions suggest irreversibility of self-assembly with distinct non-symmetric attractive and symmetric repulsive interactions. Detailed features of the dynamics become apparent when observing the acceleration (Fig. 4b) of a particle pair marked with the dotted line in Figure 4a. There are two important aspects of the interaction that can be found from this graph. First, the peak heights decrease over time indicating dissipation of energy and irreversibility. Once particles settle to an organized state with constant interparticle spacing, this condition is maintained without external disturbance. Importantly, this organized state is only achievable within a moving fluid that requires a constant external energy source (pressure gradient across the resistive channel), which is an important aspect of dynamic self-assembly which differs from static self-assembly. Secondly, acceleration patterns for repulsion and attraction are different. The first acceleration peak of particle 1 (lagging particle) and particle 2 (leading particle) are synchronous and correspond to a repulsive interaction. However, the second peaks, corresponding to an attractive interaction, are asynchronous. This off-set can be interpreted as the lagging particle first catching up followed by the leading particle slowing down. Note that this acceleration pattern repeats in the following peaks as well (Fig. 4b). These results show that fluid inertia has little effect in the repulsive interaction suggesting a unique mechanism for dynamic self-assembly consistent with the data.

Figure 2: Defocusing dynamics (a) An inertially ordered particle pair moves away from the single particle trajectory (yellow dotted line). Inset: average migration distance (N=8) (b) Schematic representation of the defocusing mechanism involving inertial lift forces (F_L) and the wall-induced reversing wake effect (F_W).

Figure 3: Reversing streamlines around spherical particles in a rectangular channel flow. The particle rotates and the wall is moving in the reference frame of the particle. (a) Finite element method simulation with Re = 0. (b) Tracer particles (1 µm) change direction near a 10 µm sphere. (Re=0.36).
The repulsive interaction initiated by a viscous wake ($F_V$), becomes strong at small interparticle spacings ($\sim O(1/l^2)$)\cite{4, 8}, (Fig. 4c). This viscous wake reflected from the nearby channel wall pushes particles off their focusing positions into staggered $y$-positions ($\Phi$). The parabolic flow distribution (gray arrows) across different $y$-positions next magnifies interparticle spacings ($\Phi$). That is, particles are pushed apart due to the parabolic velocity distribution. The results in Figure 4a shows that the leading particle moves closer to the center of the channel when it interacts in this repulsion phase. Particles however do not move off to infinity but are pushed back towards focusing positions and trajectories are stabilized by inertial lift force ($F_L$) ($\lambda$). Multiple oscillation cycles to reach focusing positions can be explained by overshooting ($\lambda \rightarrow \Phi$). Further experimental results showing an asynchronous attractive interaction in Figure 2b suggests that the lagging particle (particle 1) returns back to focusing position before the leading particle (particle 2) and this observation is consistent with the difference in inertial lift force magnitude on different sides of the inertial focusing position. The lift force becomes larger nearer to the channel wall where the lagging particle is pushed than near to the channel centerline corresponding to the position of the leading particle.

**CONCLUSION**

We have investigated dynamics of particle-particle interactions in finite-Reynolds-number channel flow. We showed that the reversing wakes reflected from channel walls are responsible for defocusing of particles and that these waves are also relevant in all microfluidic flows. The reversing wake is mainly due to confinement and does not scale with Reynolds number. Normally the particles in the channel flow simply bypass one another or interact unstably, i.e. repelled to infinity, due to viscous wakes reflected off nearby walls. Fluid inertia acts to stabilize the system keeping the particles organized at finite and precise spacings. Finally, in low Reynolds number microfluidic flows, particle separation efficiencies should be able to be improved by engineering a system that leads to particle placement and flow further from walls, where the reversing wake initiates.

**REFERENCES**