Contactless Manipulation of Microparticles by Acoustic Vortex in Droplets

Shuren Song, Jia Zhou, Antoine Riaud
State Key Laboratory of ASIC and System, School of Microelectronics, Fudan University, Shanghai, P. R. China
(Dated: March 28, 2022)

In this paper, we experimentally investigate the effect of particle diameters (20 nm, 100 nm, 450 nm, 1 μm, 5 μm and 10 μm) and liquid viscosity (0.892 mPa.s, 5 mPa.s, 18.1 mPa.s, 45.4 mPa.s and 156 mPa.s) for particle concentration and accretion by acoustic vortex with topological charge \( \ell = -15 \) in the sessile droplets. Three very interesting aggregation states are experimentally observed: Nebula, Black Hole and White Dwarf. In the experiments, we successfully concentrate particles with diameters smaller than 20 nm to a cloudy state and aggregate 5 μm and 10 μm particles to a spot. It is worth noting that we find a state of particle concentrating in most cases of our experimental conditions is a black hole will appear in the sessile droplet and the capture radius (Black Hole radius) is directly affected by particle size and liquid viscosity.

I. INTRODUCTION

Particles contactless manipulation such as concentration, aggregation and separation in microfluidics is previously considered to be the result of the combined action of the two forces: acoustic radiation force \( (F_{\text{Gov}}) \), acoustic streaming drag force \( (F_D) \) or tea leaf effect [1–3]. However, it is unlikely to be due to the radiation force because similar phenomena can be observed without acoustic field, which can reference the study of Yeo et al. [4]. In the tea leaf effect, the aggregation is driven by the secondary flow at the bottom of the tea pot (Eckmann layer) that converge to the center. Once there, the leaves are held in place by gravity, which balances the upward drag force. As pointed out by the title of Yeo et al. [4], the microfluidic version is simply an analogy. Indeed, gravity scales as \( \frac{d^2 p}{\mu} \) and the drag force scales as \( d_p \). Therefore, if the tea leaf experiment had to be repeated 10 μm particles (1000 times smaller than a small tea leaf), the density difference with water would need to be 1,000,000 times larger to hold the spheres in place. Since radiation force and gravity offer no rational explanation for SAW-driven particle concentration in sessile droplets, we propose to investigate particle dynamics in SAW-spinned droplets depending on the liquid viscosity and particle size.

Unlike previous experiments [1, 5], in this paper, we use acoustic vortices to accrete the particles in the droplets. The main advantage is that the center of the droplet is topologically protected from the incident acoustic field, with a protection radius \( r = \ell/k_0 \) (where \( k_0 \) is the wavenumber in water). This means that regardless of the scattering in the droplet, provided that axisymmetry is respected, the acoustic field at the center of the droplet is null. Using swirling SAW with topological charge \( \ell = -15 \) \( (W_{-15}, \text{the negative sign is obtained after correcting for a sign error in the formula for the transducer geometry in Ref [6])} \) at 20 MHz, the extent of the topologically protected region is approximately 180 μm radius. Any aggregation occurring within this region is guaranteed to be due to non-acoustic forces.

In this paper, we experimentally investigate the aggregating situations of particles with different diameters respectively mixed with liquids with different viscosity under acoustic streaming generated by acoustic vortices in the sessile droplets. Although the effect of viscosity had been demonstrated in both numerically and experimentally in sessile droplets excited by SAW [7], the situation of acoustic streaming generated by acoustic vortices does not be reported. Multigroup experiments are implemented utilizing the spiraling IDT with topological charge \( \ell = -15 \). According to the different states of particle aggregation, we have three classifications named: Nebula (A cloud-like cluster of particles appeared in the center), Black Hole (A black area resembling a black hole appeared in the center, surrounded by fluorescent particles) and White Dwarf (A white bright spot which fluorescent particles basically gather on this bright point appears in the center). These unique phenomena have the potential to facilitate microcentrifugation in microdroplets.

II. METHOD

The common methods to accrete particles are through generating a vortical streaming in microfluidics, including passive actuation and active actuation [8]. Passive actuations are mainly relayed on external pressure supplying equipments such as pumps to the microchip through a complex pipeline system to provide stable and continuous pressure generated, which passively generate hydrodynamic eddies in the complex geometric chamber structure [9–12]. However, the existence of pipes and large pumps squanders the miniaturization advantage of microfluidic chips. Active concentration systems, such as
FIG. 1. Experiment principle and setup. (a) Acoustic streaming generated in sessile droplets: SAW generated by interdigitated transducer leak into the liquid to form bulk acoustic wave and then transfer its momentum to the liquid to generate acoustic streaming. (b) Schematic illustration of the helix-like isophase of an acoustic vortex traveling in a sessile droplet. The incident swirling SAW is shown as an undulation at the bottom of the droplet. (c) The schematic diagram of acoustic streaming flow generated by acoustic vortex synthesized by the spiraling interdigitated transducer. (d) and (e) are phase and amplitude measured by laser doppler vibrometer of the field of the acoustic vortex \( W_{15} \) generated by spiraling IDTs. The max amplitude in (e) is approximate to 1 nm.

FIG. 2. Particles with 100 nm diameters concentration. (a) The results for 100 nm diameters fluorescent particles concentration when turning the swirling SAW off and on. (b) Schematic diagram of light path convergence. (c) and (d) Line and bar graphs of fluorescence intensity.

electro-kinetics [13], thermal actuation [14–16], MEMS [17, 18], and acoustofluidics method [19], especially the latter, have little or no requirements for tubings and pumps and more suitable for the integrated development of microchips. Straight IDTs are suitable for particle aggregation due to the ability to generate a continuous and stable acoustic field which can asymmetrically leak into sessile droplets to generate a vortex flow. Nonetheless, unidirectional and single acoustic field results in unstable acoustic streaming in the sessile droplet, which significantly limits application of the IDTs in microcentrifugation. Similar to the IDTs design in Ref [6, 20], we
FIG. 3. Three classification results: (a) and (d) Nebula, (b) and (e) Black Hole, (c) and (f) White Dwarf for different size red fluorescent particles manipulated by acoustic vortex in water. Here, each picture is the superimposed average value of 200 frames of image pixels before the SAW is turned off in the video. The input voltage during the experiments is 200 mV. (g) is obtained by intercepting a picture every 10 seconds of the above experiment videos and arranging them in time sequence.

prepared spiraling IDTs which can generate $W_{-15}$ in this section.

A. Experiment principle

The cyclone-like flow in the droplet is driven by the acoustic streaming of an acoustic vortex. The helical wavefront of such screw dislocation is shown in Fig. 1 (b). The droplet diameter (2 mm) is much larger than the acoustic wavelength (75 µm) at the acoustic frequency used in this work (MHz). We consider a fluid of density at rest $\rho_0$, kinematic viscosity $\nu$, kinematic bulk viscosity $\nu'$ and sound velocity $c_0$ subject to an acoustic field of angular frequency $\omega$, wavenumber $k_0$, pressure $\tilde{p}$ and oscillating velocity $\tilde{v}$. The kinematic bulk viscosity represents the non-shear irreversibilities observed when compressing a fluid (such as during an acoustic wave propagation) and is zero for monoatomic gases [21, 22]. Analytical expressions of the acoustic streaming field of steady velocity $\vec{v}$ and steady vorticity $\vec{\Omega}$ can be derived by perturbation expansion when the acoustic pressure is not too large ($\tilde{p} \ll \rho_0 c_0^2$) and the acoustic attenuation is moderate $(\frac{2\omega c_0^2}{\nu} \gg 1)$, where $b = \frac{4}{3} + \nu'/\nu$. This allows successively solving fluid quantities of a field $x$ first with zero-order rest quantities $x_0$, then as small first-order perturbation expansion acoustic quantities $\tilde{x}$ and finally very small second-order steady state effects of nonlinear acoustics $\bar{x}$. Upon such perturbation, acoustic streaming can be modeled as a steady solenoidal acoustic streaming force $\vec{F}$ driving a Stokes flow [7, 23]:

$$\nu \nabla \times \vec{\Omega} = \vec{F}, \quad (1a)$$

with:

$$\vec{F} = \frac{\omega^2 \nu b (\Pi)}{c_0^2}, \quad (1b)$$
FIG. 4. The experiment results for red fluorescent particles with diameter equal to: 20 nm, 100 nm, 450 nm, 1 µm, 5 µm, 10 µm actuated by acoustic vortex in aqueous glycerol solutions of different viscosities: 0.892 mPa.s, 5 mPa.s, 18.1 mPa.s, 45.4 mPa.s, 156 mPa.s. Here, each picture is the superimposed average value of 200 frames of image pixels before the SAW is turned off in the video. The input voltage during the experiments is 200 mV. Among them, a blue box and a green box are used to distinguish the aggregation state of these results, a yellow box represents the unstable status, and red box represents the unstable state in dark hole.

where \( \langle \Pi \rangle \) indicates the time-average of the acoustic power flux \( \Pi = \bar{p} \bar{v} \), which is also a second-order perturbation. We note that while acoustic streaming can develop into high-Reynolds number turbulent jets [24], the theory is rigorously tractable only for creeping flows as above. Quite interestingly, the acoustic streaming force is set only by the acoustic field, which can be precisely controlled in three dimensions by holographic techniques [25–27]. By using the acoustic streaming to generate a steady flow, and then turning the acoustic field off, one can follow the spontaneous (non-forced) evolution of an arbitrary flow structure. Conversely, one can keep the acoustic forcing on at all times to generate a steady flow as it will be done in the remaining of the paper. The first-principle simulations results based on Eq. (1b) and Eq. (4) were carried out, the simulation results shown that the direction of acoustic streaming generated by \( W_{-15} \) in the droplet center axis was spiraling downward, which can be regarded as an attractor streaming (the acoustic streaming is pulled to the transducer, see Fig. 1 (c)) [25].

B. Acoustic vortex

Acoustic vortices can be generated by various means, for instance, diffraction networks [28], using an acoustic source with a helicoidal radiating surface [29], via the optoacoustic technique [30], metamaterials [31, 32], transducer arrays [25, 33, 34] and spiraling IDTs patterned on a piezoelectric substrate [6, 35]. For facilitating the design and the integration process simplify, we choose the latter approach.

A classical solution to the wave equation in isotropic medium is known as the Bessel beam [25]:

\[
W_t e^{-i\omega t} = J_\ell(k_r r) e^{i\phi + ik_z z - i\omega t},
\]

(2)

\[
W_0 e^{ik_z z - i\omega t},
\]

(3)

Where \( r, \theta, z, t, \ell, W_t, J_\ell, k_r, k_z, \omega, \) and \( W_0 \) denote, respectively, for the axisymmetric coordinates, the time, the topological order, the complex wave-field value, the \( \ell \)th-order Bessel function, the radial and axial parts of wave vector, the angular frequency, and the isotropic swirling surface acoustic wave complex value. Then we get the isotropic acoustic vortex:

\[
W_0^0(r, \theta) = \frac{1}{2\pi i t} \int_{-\pi}^{+\pi} e^{i\ell \phi + ik_z r \cos(\phi - \theta)} d\phi,
\]

(4)

By analogy with the isotropic equation, we define an anisotropic swirling wave with a given \( \kappa_z = k_z \) as:

\[
W_\ell^0(r, \theta) = \frac{1}{2\pi i t} \int_{-\pi}^{+\pi} e^{i\ell \phi + ik_r (\phi, k_z) r \cos(\phi - \theta)} d\phi,
\]

(5)
FIG. 5. The trajectory diagram of 10 µm fluorescent particles in liquids of different viscosities with different input voltages. Here, each picture is the superimposed average value of 200 frames of image pixels before the SAW is turned off in the video. The X-axis of the Sudoku is the different input voltages: 200 mV, 300 mV and 400 mV. The Y-axis of the Sudoku is the different viscosity: 18.1 mPa.s, 45.4 mPa.s and 156 mPa.s.

C. Spiraling interdigital transducer

Based on the Eq. (5) and Ref [6], we design the spiraling IDT and the preparation process is as follows. We firstly design a mask of the spiraling IDT. Then, with 10 nm titanium (Ti) as the adhesion layer and 100 nm gold (Au) as the electrode, the spiraling IDTs are deposited on the 1 mm thickness X-cut lithium niobate by Physical Vapor Deposition (PVD). The electrode patterns finally are gotten by standard ultraviolet (UV) light photolithography process and wet etching (see Fig. 1 (c)). In addition, phase and amplitude (see Fig. 1 (d) and (e)) of $W_{-15}$ were measured by laser Doppler vibrometer (LDV) to accurately determine the properties and performance of the spiraling IDT. Here, the maximum amplitude of $W_{-15}$ approximately 1 nm with 200 mV (peak to peak) input voltage to the signal generator (see Fig. 1 (e)). The reason to chose swirling SAW $W_{-15}$ to study particles concentration is that the dark core which can be found in the center of the swirling SAW size is equal to the droplet radius (volume: 2 µL). This ensures that there is no acoustic wave at the center of the droplet, which allows to eliminate the effect of radiation pressure in the data analysis. Moreover, the relevant experimentally study of the acoustic vortex with a large topological charge ($\ell > 10$) has not been reported recently.

D. Experiment preparation

Before the experiments, a 2 mm diameter gold ring with 100 µm width is designed (see Fig. 1C) which the surface is hydrophobically treated with dodecyl mercaptan (1-dodecanethiol, SIGMA, USA) to ensure the droplet is accurately located in the center of the transducer. During the experiments, 2 µL sample mixture droplet with fluorescent particles is dropped onto the ring range through a pipette each time to ensure the consistency of the experiments. Here, we prepare glycerol aqueous solutions with different viscosities $\mu$: 0.892 mPa.s (DI water), 5 mPa.s, 18.1 mPa.s, 45.4 mPa.s, 156 mPa.s. Meanwhile, red fluorescent polystyrene microspheres (excitation wavelength: 535 nm, emission wavelength: 610 nm, HUGE, Shanghai, China) with different diameters $d_p = 20$ nm, 100 nm, 450 nm, 1 µm, 5 µm, 10 µm are dropped in the above solutions and preserve sealed at room temperature. Then a signal generator (MFG2260M, CN) in series with the amplifier (ZHL-5W-1+, USA) and input sinusoidal signals with frequency $f_0 = 20$ MHz, peak-to-peak voltage from $V_0 = 60 \sim 400$
mV to the spiraling IDTs. A high-speed camera (Basler acA720-520 µm, GER) with 500 fps is used to accurately capture particles trajectory images. In addition, ImageJ (1.53j, USA), an image processing software is used to process the videos.

After the preparatory work is completed, 100 nm fluorescent particles are aggregated with different input voltages to determine the suitable input power which can concentrate particles to form the brightest spot. During the experiments, we notice that before the concentration, the fluorescent light will be reflected at the air-liquid surface within the droplet and then converge to form a bright spot (see Fig. 2 (a) and (b)). Hence, in order to accurately obtain the effect of particle aggregation, we use the difference between the fluorescence intensity after aggregation and before aggregation to compare, the results show that the brightest spot of 100 nm particles will be formed when the input power at 210 mV (see Fig. 2 (d) and (d)). Since the voltage around 210 mV has little effect on the results, we chose the input power at 200 mV to simplify the calculating and comparing.

III. RESULTS

A. Three different concentration outcomes

First, we experimentally investigated the accretion of particles with different diameters ($d_p = 20$ nm, $100$ nm, $450$ nm, $1$ µm, $5$ µm, $10$ µm) in low viscosity (0.892 mPa.s, deionized water) by swirling SAW $W_{-15}$. When a sinusoidal signal with 200 mV (peak to peak) voltage applied to the spiraling IDT, three attractive states appeared in the sessile droplet center: (i) the cloud state named, Nebula (see Fig. 3 (a) and (d)), (ii) a bright cluster with a dark hole in the center named, Black Hole (see Fig. 3 (b) and (e)), and (iii) a bright aggregation dot named, White Dwarf (see Fig. 3 (c) and (f)). Here, experimental pictures are intercepted every 10s according to the time series for obtained video of each group.
FIG. 7. (a) The rotation speed $\omega$ for particles with diameter: 1 $\mu$m, 5 $\mu$m and 10 $\mu$m manipulated by acoustic vortex in different liquid viscosity. (b) Capture radius for different size particles (20 nm, 100 nm, 450 nm, 1 $\mu$m, 5 $\mu$m and 10 $\mu$m) manipulated by acoustic vortex in different liquid viscosity (0.892 mPa.s, 5 mPa.s, 18.1 mPa.s, 45.4 mPa.s and 156 mPa.s). (c) The schematic for the generation reason of the dark hole. (d) The calculation results for relationship between the radius of the dark and the ratio of particle size to hydrodynamic boundary layer. Here, the colorbar is the particle diameter (log10($d_p$)), the y-axis is the dark hole radius, the x-axis is the ratio of particle size to hydrodynamic boundary layer.

of experiments, which form a picture matrix of a total of 100 s (see Fig. 3 (g)). The above three states are generated from a short time (within 10 s) after opening the acoustic field and continue until the acoustic field is closed (about 100 s), which demonstrate the three states (Nebula, Black Hole, White Dwarf) are stable in the low viscosity droplets situation under the action of swirling SAW. Moreover, large particles ($d_p = 5$ $\mu$m and 10 $\mu$m) can be successfully aggregated by $W_{-15}$ to a point, while small particles (diameters smaller than 5 $\mu$m) can only be concentrated and a dark hole appear in the droplet center.

Then, to further clarify the above phenomena, we implemented multi-group experiments to aggregate particle with six size diameters (20 nm, 100 nm, 450 nm, 1 $\mu$m, 5 $\mu$m, 10 $\mu$m) mixed with the mixture of glycerin and water with five viscosity ($\mu = 0.892$ mPa.s, 5 mPa.s, 18.1 mPa.s, 45.4 mPa.s, 156 mPa.s). Here, we select the last 200 frames of the videos (each experiment video has a total of 5402 frames with 50 fps) to get the average value to obtain the clear final patterns (see Fig. 4). From the results, we find that Black Hole (in red box) is the more general case in most experiments. Although there are a dark holes can be found in the yellow box (see Fig. 4), in some instances, unstable aggregates can appear inside the dark holes. Dark holes appear like some intermediate state between aggregates and cloud, and therefore will be studied more extensively.

Moreover, the experiments for different input voltage ($V_{dp} = 200, 300, 400$ mV) applied to the spiraling IDTs to aggregate particles diameters $d_p = 10$ $\mu$m in high viscosity liquid ($\mu = 18.1$ mPa.s, 45.4 mPa.s, and 156 mPa.s) are implemented to investigate the effect of input power and viscosity for the particle aggregation. To facilitate comparison, we also select the last 200 frames of the videos for the entire process and get the average value to obtain the droplet pattern. As shown in Fig. 5, Black holes still appear in the center of the droplet with 10 $\mu$m particles under different input voltages (200 mV, 300 mV, 400 mV) and high viscosity (18.1 mPa.s, 45.4 mPa.s and 156 mPa.s), which we infer that the dark hole should be the norm status during particle aggregation. Here an interesting but complex phenomenon occurred that bright spots will appear randomly in the center of the dark hole (Fig. 5 (d) and (e)) similar to the results of yellow box in Fig. 4. Different with 200 mV voltage situation, the droplets will become unstable with higher input voltages (300 and 400 mV) as is expected even with high viscosity, which makes the profile of the dark holes become blurred (see Fig. 5 (c), (f) and (i)).
FIG. 8. (a) The relationship between the force subjected to the particles and Reynold number. The blue stars are cloud, the orange discs are dark hole, the green dots are aggregates. (b) Postulated aggregation mechanism. The deviation must be strong enough so that the particle moves from its original streamline into the attraction basin. Note that the starting height is controlled by the particle size, and therefore larger particles have a longer distance to travel to reach the attractor basin. (c) Spatio-temporal cross-sections of 5 \( \mu \)m and 10\( \mu \)m particles aggregated in water (0.892 mPa.s)

The same phenomenon also appear in the experiments of 5 \( \mu \)m particles which are aggregated by different input voltages in high viscosity (156 mPa.s) (Fig. 6 (a), (b) and (c)). It’s worth noting that not only dark holes will appear in the sessile droplet center, but also the aggregation of particles in the dark hole center sometimes. In order to clarify the state of particles gathering at the center of the dark hole, we unfold the entire video in a time series sequence and get the spatio-temporal cross-sections (see Fig. 6 (d) and (e)). Here, lifetime \( \tau \) represents the time of particles aggregation which a bright area enclosed by the yellow box in the droplet center (see Fig. 6 (d) and (e)). The length of lifetime \( \tau \) shown in Fig. 6 (d) and (e), is irregular which proves the high input power cause complex acoustic streaming field in the sessile droplet and droplets vibration which induced the particles’ motion irregular.

B. The explanation for dark holes

Due to the droplet vibration when apply the high input power, we select the velocity of the acoustic streaming flow at the center of the droplet, where the rotation is relatively stable, to determine the rotating speed \( \omega \) of the droplet. Obviously, the particles size has little effect to the rotating speed of the droplet and \( \omega \) will decrease as the viscosity of the liquid increases from Fig. 7 (a).

In addition, we measure all the capture radius of particles size (20 nm, 100 nm, 450 nm, 1 \( \mu \)m, 5 \( \mu \)m and 10 \( \mu \)m) during the experiments in different liquid viscosity (0.892 mPa.s, 5 mPa.s, 18.1 mPa.s, 45.4 mPa.s and 156 mPa.s) shown in Fig. 7 (a). We find that the dark hole radius increases with particle size increasing and decreases with liquid viscosity increasing. An obvious trend is described in Fig. 7 (b): the capture radius, that is, the radius of the dark hole will display with different changing range. When the particle diameter is small (20 nm and 100 nm), the capture radius (that is, the radius of the dark hole) is very stable close to zero, which can be regarded as Nebula. We postulate that the holes are formed via a purely steric mechanism. Particles are carried to the center of the droplet via some toroidal streamlines (see Fig. 7 (c)). The closer the streamline to the axis of the droplet, the closer it also flows next to the solid substrate. Therefore, large particles cannot flow along the streamlines closest to the center of the droplet. If such mechanism is true, then the dark hole radius should only depend on the droplet radius and the ratio of the particle size to the boundary layer thickness. The boundary layer is estimated as the minimum between half the droplet radius and the Blasius boundary layer thickness, with the distance \( x \) taken as half the droplet radius:

\[
\delta(x) \approx 5.0 \sqrt{\frac{\nu x}{u_0}} = 5.0 \frac{x}{\sqrt{Re x}}, \tag{6}
\]

We calculate the trend by Eq. (6) as shown in Fig. 7...
(d), which indicates that the boundary layer effects the particles and hinders the particles to go to the droplet center and then the dark holes appear.

C. The explanation for particles aggregation

Tracing the resulting state depending on the Reynolds number (Re) and the force $F_p$ on the particle, we get Fig. 8 (a). Interestingly, all the 5 and 10 $\mu$m particles feature aggregates with various degrees of stability. Spatio-temporal cross-sections are shown in Fig. 8 (c).

We find that 5 and 10 $\mu$m particles are consistently focused at the center (V-shape). This indicates that some force is able to attract particles towards the center. Because the axis of the droplet is in the hollow region (which is topologically protected), only hydrodynamic forces can explain the migration. Based on Stokes-Boussinesq-Oseen, the only attractive force can be the pressure gradient. Furthermore, assuming that the particles move according to the gradient of the pressure, the accumulation of particles (div(grad)) is given by the Laplacian of the pressure. Based on the works of Douady et al. [36], the pressure Laplacian is proportional to the square of the vorticity minus the square of the shear. For $\Delta = 3\pi d\mu$, the pressure Laplacian is proportional to

$$
\nabla^2 p = \frac{3\pi d\mu}{2}
$$

Within a time $\delta t = \frac{R_d}{v_p}$ and a distance $L \simeq R_d$.

From the relationship

$$
\dot{r} \delta t > \delta r,
$$

we get:

$$
d_p > \frac{18 \beta v_p \mu}{R_d \Delta \delta t}
$$

where $\Delta = \frac{\alpha^2 \pi}{4\beta}$.

The model assumes that the radial position of the particle before it meets the attractor is related to its position in the boundary layer. The outcome of the deviation due to pressure forces is decided based on a kinematic condition (“distance to cross”). Based on the important role played by the boundary layer in structuring the dark hole, and the postulated appearance of an eddy, we postulate the following mechanism for aggregation: particles are always attracted. Depending on their starting point and migration speed, they may (i) end up in the basin of attraction (aggregate) (ii) be deviated outward their original streamline and end up on a streamline that goes too close to the solid surface, so that the particle bottom sticks to the solid (>5 mPa.s case). If the viscosity is large enough to detach the particles, or deviation is not too big, they may come back later (>18.1 mPa.s).

In other words, for a particle to aggregate, the Reynolds number must be large enough to form an eddy, then the attraction must be strong enough to capture the particle at the first attempt, otherwise it escapes and may stick to the surface of the droplet (low viscosity) or cycle back.

D. Seed-assisted aggregation

Due to the existence of the Black Hole, $W_{-15}$, even can be used to separate particles with different size diameters which are uniformly distributed in sessile droplets (2 $\mu$L, see Fig. 9). An very interesting result shown that red fluorescent particles (Diameter: 10 $\mu$m) are coincidently aggregated in the center of dark holes formed by blue fluorescent particles (Diameter: 1 $\mu$m) whereas 1 $\mu$m particles alone could not be aggregated.

IV. CONCLUSION

In this paper, we mainly investigate particle accretion under the contactless acoustic streaming driven by $W_{-15}$ in the sessile droplet. We experimentally observe three complex states, namely: Nebula, Black Hole and White Dwarf. The reason for these phenomena appearing is mainly relative to the particle diameter and liquid
viscosity, which change the Reynold number of the liquid and limit the moving distance of particles with different size to go to the center. During the experiments, we notice that the droplet vibration actually has little influence on the concentration of the particles. We successfully aggregate 5 μm and 10 μm particles to a bright spot and realize different size particles separation. Different from the former studies, we investigate the influence of the liquid viscosity from low viscosity (0.892 mPa.s) to high viscosity (156 mPa.s) and prove that the swirling SAW with large topological charge ℓ can also be used to concentrate particles. Moreover, 10 μm particles can be used as seeds to aggregate 1 μm particles. About the minimum sorting size or mechanism does give a migration condition that is more easily satisfied for large particles (at any given flow pattern). In the future, additional simulations are needed to confirm the present postulated mechanism.

[1] Ghulam Destgeer, Hyunjun Cho, Byung Hang Ha, Jin Ho Jung, Jinsoo Park, and Hyung Jin Sung. Acoustofluidic particle manipulation inside a sessile droplet: four distinct regimes of particle concentration. *Lab on a Chip*, 16(4):660–667, 2016.
[2] Tao Peng, Cui Fan, Mingyong Zhou, Fengze Jiang, Dietmar Drummer, and Bingyan Jiang. Rapid enrichment of submicron particles within a spinning droplet driven by a unidirectional acoustic transducer. *Analytical Chemistry*, 93(39):13293–13301, 2021.
[3] Jingui Qian, Wei Huang, Renuhua Yang, Raymond HW Lam, and Joshua E-Y Lee. Low-cost laser-cut patterned chips for acoustic concentration of micro-to nanoparticle and cells by operating over a wide frequency range. *Analytical Chemistry*, 146(10):3280–3288, 2021.
[4] Leslie Y Yeo, James R Friend, and Dian R Arifin. Electric tempest in a teacup: The tea leaf analogy to microfluidic blood plasma separation. *Applied Physics Letters*, 89(10):103516, 2006.
[5] Anliang Zhang, Weiyue Liu, Zhidi Jiang, and Jingchen Fei. Rapid concentration of particle and bioparticle suspension based on surface acoustic wave. *Applied Acoustics*, 70(8):1137–1142, 2009.
[6] Antoine Riaud, Michael Baudoing, Olivier Bou Matar, Loic Becerra, and Jean-Louis Thomas. Selective manipulation of microscopic particles with precursor swirling Rayleigh waves. *Physical Review Applied*, 7(2):024007, 2017.
[7] A Riaud, M Baudoing, O Bou Matar, J-L Thomas, and P Brunet. On the influence of viscosity and caustics on acoustic streaming in sessile droplets: an experimental and a numerical study with a cost-effective method. *Journal of Fluid Mechanics*, 821:384–420, 2017.
[8] Heba Ahmed, Shwathy Ramesan, Lillian Lee, Amgad R Rezk, and Leslie Y Yeo. On-chip generation of vortical flows for microfluidic centrifugation. *Small*, 89(10):103516, 2006.
VII. ADDITIONAL INFORMATION

Supplementary information is available for this paper at XXX.