Hydrodynamically induced rhythmic motion of optically driven colloidal particles on a ring

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We experimentally study the motion of optically driven colloidal particles on a circular path by varying their number N. Although an identical driving force is applied to each particle, their equally spaced configuration is hydrodynamically unstable, and a doublet configuration is spontaneously formed. In small-N systems, the angular difference between neighboring particles exhibits oscillatory or nonoscillatory behavior. The number of oscillatory modes that appear depends on the maximum number of doublets that the system can contain. Frequent switching between different modes was observed with increasing N. The characteristic frequencies of the oscillatory modes are discussed theoretically by linear stability analysis of the equations that govern the motion of hydrodynamically coupled particles. The evaluated frequencies of the slowest modes exhibit reasonably good agreement with those of the mainly observed modes in experiments. The relationship between the characteristic frequencies and specific configurations is confirmed experimentally by setting a specific initial configuration for the particles. An increase in N also enhances the mean angular velocity of the particles owing to the reduced effective viscosity in large-N systems.

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I. INTRODUCTION

Self-propelled micro-objects have recently attracted the attention of researchers as typical examples of microscopic nonequilibrium systems. At the mesoscopic scale, the Reynolds number becomes small, and the hydrodynamic interaction plays an important role in their dynamics [1–3]. The hydrodynamics for a low Reynolds number is well described by the Stokes approximation with the Oseen mobility tensor [1]. Micro-objects are often found to exhibit novel collective motion owing to their long-range hydrodynamic interaction. Since this hydrodynamic interaction is symmetric for the exchange of particles, it has no effect on the motion of equally or identically driven particles. However, there are some situations where the asymmetric motion of particles is expected to occur. For instance, some microorganisms exhibit rhythmic motion near boundary layers [4], sediment particles in a viscous flow exhibit chaotic behavior [5], and an acoustic phonon propagates in an array of flowing droplets confined in a thin cell [6]. Studies on such behavior are also important to explore the physical mechanism of the swimming and pumping of living micro-objects.

An optical manipulation technique involving optical tweezers has frequently been utilized to realize mesoscopic multi-particle systems in a controlled manner [7–13]. A particle optically driven by this method can be regarded as a minimal model of self-propelled particles. When particles are confined to a circular path, there is no limitation of the system size along the path, and the hydrodynamic coupling between the particles becomes stronger due to the finite distance between the particles. A system composed of three particles has been reported to display a limit cycle [14–16] and chaotic behavior [14]. However, the collective motion in a multiparticle system, where the many-body effect or crowding effect is expected to play a crucial role, is still an open question.

II. EXPERIMENT

In this paper, we report various types of characteristic rhythmic motion of optically driven particles on a circular path for different numbers of particles N. The number of types of characteristic motion is determined by the maximum number of doublets that the system can contain. The relationship between the characteristic motion and the specific configuration of the particles is studied experimentally by setting a specific initial configuration for the particles. The characteristic modes are also discussed theoretically by linear stability analysis of the equations of motion for hydrodynamically coupled particles. The dependence of the average velocity of the particles on N is also discussed by considering the characteristic collective motion of the particles.
objective lens (Plan Fluor, Nikon, 100×, NA=1.4). This lens was used to project a ring-vortex trap in a sample cell and to image the particles. A dichroic mirror (Sigmakoki) was used to reflect the laser beam and to observe images under visible illumination. We used a ring-vortex trap with radius $R = 10 \mu m$ and topological charge $l = 45$. The power of the laser beam was fixed to 2.0 W during the experiment. The images were captured by a monochrome CCD camera (ADT-100, Flovel, 1000 × 1000 pixel$^2$, 10 bits) at 30 frames/s. The instantaneous positions of the particles were extracted using digital video analysis with IMAGEJ.

Spherical silica particles of 3.0-μm diameter (Hipersica, UNK) were dispersed in an 85-μm-thick layer of water between cover slips. Since the particles were stored in a mixed-bed ion-exchange resin for a month before use, they were expected to be highly charged. The circular path was set about 10 μm above the bottom of the cell to reduce hydrodynamic coupling to the cell wall. The sample cell set about 10 μm-thick layer of water above the bottom of the cell to reduce noise. From the slope of $\theta_i(t)$, as shown in Fig. 2.

In the $N = 1$ system, the angular distance $\theta_1(t)$ increases almost linearly with the elapsed time $t$, as shown in Fig. 2. The mean velocity of the particle is about 10.0 μm/s (1.00 rad/s) and is much larger than the diffusion length of the particle per second. Although the displacement of the particle due to thermal fluctuation is small compared with the displacement induced by the driving force in our system, there is some fluctuation in $\theta_1(t)$. This is due to the nonuniformity of the optical driving force along the circular path [14]. The fluctuation in velocity of the particle in the $N = 1$ system is about 27%. This fluctuation in the driving force can be regarded as nonthermal (quasiperiodic) noise in our experiment.

In the $N = 2$ system, the slope of the angular distance of particle $\theta_2(t)$ changed at approximately $t = 22.5$ s, as shown in Fig. 2. The angular difference $\Delta \theta_2(t)$ decreases for $t \leq 22.5$ s, after which it remains at about 0.51 rad. This indicates that the particles form a doublet without direct contact, and this configuration is hydrodynamically stable against nonthermal noise. From the slope of $\theta_2(t)$ for $t \geq 22.5$ s in Fig. 2, the mean velocity of the doublet in the $N = 2$ system is about 13.2 μm/s. A doublet moves faster than a single particle. This is due to the reduction of effective viscous drag for the backward particle, which originates from the mutual hydrodynamic interaction between the particles in this symmetry-breaking situation. The tendency that a doublet moves faster than a singlet is a general feature also observed in $N \geq 3$ systems.

The stable angular difference $\Delta \theta_2(t)$ of 0.51 rad corresponds to a center-to-center distance of 5.0 μm. Thus, there is a gap between the particles. This suggests that there have to be some attractive and repulsive forces that maintain the stable interparticle separation. Doublet formation in a similar system was recently studied in detail by experiment and simulation [22]. In a general situation, since the hydrodynamic forces between two particles cancel out, the particles do not always approach each other. However, when they are confined to a circular path by a finite bounded force, the radial component...
of the hydrodynamic force shifts the relative radial positions of the particles. This causes the velocity of the front particle to decrease and that of the backward particle to increase. Therefore, the backward particle catches up with the front particle irrespective of their initial angular positions.

There are various possible origins of the repulsive force. Since the silica beads used in this study are highly charged in a low ionic solution, one possible origin is electrostatic repulsion [16]. The distance at which the electrostatic repulsive interaction is greater than the thermal energy is as large as micrometers [23]. According to the histogram of $\Delta \theta_2$, the minimum value of $\Delta \theta_2$ is 0.4 rad, and the corresponding interparticle distance is about 3.97 $\mu$m. This value can be explained by the electrostatic interaction alone, even if the interaction is screened to some extent. However, we cannot explain the average value of 5 $\mu$m by the electrostatic interaction alone. Since the distance between the surfaces of the particles is as small as the radius of the particles, the hydrodynamic lubrication effect may be another possible origin of the repulsive force. This dynamical contribution increases the stable interparticle distance. In addition, when two particles approach each other, the radial component of the hydrodynamic force decreases, and the difference in the velocity of the particles decreases. This reduces the attractive force considerably, and the stable interparticle distance tends to increase owing to fluctuation.

**IV. RHYTHMIC MOTION OF PARTICLES IN $N \geq 3$ SYSTEMS**

According to a theoretical study [15], the equally spaced configuration in our system is unstable owing to the hydrodynamic interaction between the particles. The particles are expected to exhibit some stationary or rhythmic motion in many-particle systems.

In the $N = 3$ system, the configuration observed in the experiment is composed of one doublet and one single particle, as schematically shown in Fig. 3(a). This configuration induces a limit cycle, which has been shown experimentally [14,16] and theoretically [15]. Once a doublet is formed, it accelerates and catches up to the preceding single particle [Fig. 3(a), region A]. They then form a transient cluster of three particles [triplet; Fig. 3(a), region B]. However, the middle particle in the triplet is subjected to less viscous drag than the others owing to the reduced effective viscosity [1,15]. Therefore, the front two particles move at a greater velocity [Fig. 3(a), region C] and catch up to the single particle from behind again. This periodic approach and escape of a doublet with the exchange of the particles in the doublet is repeated.

In Fig. 3(b), this periodic collective motion can be observed as the oscillation of $\Delta \theta_3$. Although the limit cycle eventually collapses at approximately $t = 735$ s, it immediately recovers. This indicates that a doublet is more stable than two singlets. The frequency spectrum of $\Delta \theta_3$ is also shown in Fig. 3(c). The characteristic frequency of the oscillation observed in Fig. 3(b) is 16.7 mHz.

In the $N = 4$ system, oscillatory motion only appears for $t \leq 500$ s [region A in Fig. 4(a)]. After that, the oscillatory motion disappears [region B in Fig. 4(a)]. The periodic oscillation in region A corresponds to a configuration composed of one doublet and two singlets. The exchange of the particles in the doublet causes the oscillation in $\Delta \theta_4$, similar to that in the $N = 3$ system. On the other hand, only a steady configuration with two independent doublets is observed in region B for $t > 500$ s, where the motion of the two doublets does not exhibit periodic oscillation in $\Delta \theta_4$. The continuous small values (0.55 rad) of $\Delta \theta_4(1,2)$ and $\Delta \theta_4(3,4)$ indicate the stability of the doublets.

If we regard each doublet as a single particle, the formation of a pair of doublets is expected to be observed. When the two doublets approach each other, the following process is observed: the front particle in the backward doublet accelerates, causing the backward doublet to break into two singlets. This reduces the velocity of the front particle of the backward two singlets, and the front doublet moves away. Then, the backward two singlets form a doublet again. Therefore, a stable triplet or quartet is not observed in the $N = 4$ system. Only a large fluctuation between the two doublets is observed, as shown by the plots of $\Delta \theta_4(2,3)$ and $\Delta \theta_4(4,1)$ in Fig. 4(a).

The frequency spectrum of $\Delta \theta_4$ is shown in Fig. 4(b). The characteristic frequency of the oscillation is 35.8 mHz. This frequency corresponds to that of oscillation in region A. The small peaks around 200 mHz correspond to the small oscillation in region B.

The motion observed in our systems can generally be classified into two types, oscillatory motion and steady configuration motion, which can be distinguished from each other by the temporal changes in $\Delta \theta_8$. The former is observed...
in a configuration composed of doublets and singlets and originates from the difference between the velocity of a singlet and a doublet. The reorganization of a doublet causes $\Delta \theta_N$ to oscillate with time. The latter is only observed in the even-$N$ systems. When the particles exhibit this motion, only doublets appear in the system. Since the doublets do not exchange particles during this motion, the doublets are stable, and $\Delta \theta_N$ does not undergo a large oscillation.

In the $N = 5$ system, only oscillatory motion is observed, and a configuration consisting of two doublets and a singlet is mainly observed in our experiment. However, there is another oscillatory mode with a short time period. Some time points where such oscillation appears are marked by downward arrows in Fig. 5(a). This mode consists of one doublet and three singlets. These two modes are also observed in the frequency spectrum shown in Fig. 5(b). Peak 1 at 17.8 mHz corresponds to the former mode, and peak 2 at 57.8 mHz corresponds to the latter mode. Their relative magnitude reflects the relative appearance probability of the two modes.

In the $N = 6$ system, both oscillatory (regions A, B + A, and B) and steady configuration motion (region C) are observed in different time regions, as shown in Fig. 6(a). There are two different types of oscillatory mode typically observed in regions A and B. The oscillatory motion in region A includes one doublet, and that in region B includes two doublets. In region B + A, a mode similar to that in region B appears at an earlier time, and a mode similar to that in region A appears at a later time. The frequent switching between the oscillatory and steady configuration motion in the $N = 6$ system indicates that the steady configuration motion is less stable than that in the $N = 2$ and 4 systems. This is due to the enhancement of the hydrodynamic interaction between distant particles with increasing particle density on the circle.

The frequency spectrum of $\Delta \theta_6$ is shown in Fig. 6(b). There are characteristic peaks at 39.2, 63.3, and 216 mHz that are respectively labeled peaks 1, 2, and 3 in Fig. 6(b). We regard peaks 1 and 2 as corresponding to the characteristic frequencies of modes B and A, respectively, by comparing the local frequency spectrum in both regions. Peak 3 corresponds to the small and rapid fluctuation observed in region C, similar to that in the $N = 4$ system.

The temporal changes in $\Delta \theta_7$, $\Delta \theta_8$, and $\Delta \theta_9$ for a certain pair of particles are shown as examples in Fig. 7. Although the odd-even effect discussed above is also observed in these cases, the time period in which a certain characteristic mode exists becomes short when $N = 8$ and 9. This switching causes the temporal rhythmic motion to become more complex in larger-$N$ systems, making it difficult to clearly distinguish between the different types of motion. The frequency spectrum also becomes more continuous.

As a result of the frequent switching between the characteristic types of motion, the formation of a transient large cluster is enhanced. As a general trend, a doublet is stable and a larger cluster is unstable in our system; even if a larger cluster is formed, it soon breaks into singlets and doublets. Although the appearance probability of doublets is large even in large-$N$ systems, the probability of a transient larger cluster...
V. STABLE COLLECTIVE MOTION APPEARING IN N-PARTICLE SYSTEMS

The number of oscillatory modes is theoretically expected to increase with $N$ [15]. Systems with an odd number of particles have $(N - 1)/2$ independent oscillatory modes. On the other hand, systems with an even number of particles have $(N - 2)/2$ oscillatory modes and one nonoscillatory mode. To identify the correspondence between a specific configuration and a characteristic pattern of $\Delta \theta_n$, we set the initial configuration of the particles to the more frequently observed configuration. In our experiment, the switching between these modes is observed.

An example of the results of such an experiment for $N = 6$ is shown in Fig. 8. In the $N = 6$ system, two types of oscillatory motion and one steady configuration motion are expected to be observed. We set the following three configurations as the initial configurations, as shown in Fig. 8: (A) one doublet and four singlets, (B) two doublets and two singlets and (C) three doublets. Configurations A and B exhibit oscillatory motion in $\Delta \theta_6$, whereas configuration C exhibits steady configuration motion. The small oscillation in Fig. 8 appears to increase. The formation of a cluster is considered to occur when the angular difference between particles becomes smaller than 0.6 rad, which is determined by a histogram of $\Delta \theta_N$ for each $N$ that includes the fluctuation of each particle. The probabilities of a particle belonging to a doublet, triplet, and quartet cluster are respectively 0.35, 0.25, and 0.20 in the $N = 9$ system. These values are much larger than those in smaller-$N$ systems; for example, they are 0.48, 0.02, and 0.00, respectively, in the $N = 5$ system. In addition, the decrease in the mean particle separation should also enhance the formation of hydrodynamically unstable large clusters in larger-$N$ systems.

FIG. 6. (Color online) (a) Temporal changes in $\Delta \theta_6$. Regions A, B + A, and B [yellow (light gray)] are oscillatory motion. Region C (white) is steady configuration motion. (b) Averaged frequency spectrum of $\Delta \theta_6$. The frequency of the highest peak, labeled 1, is 39.2 mHz. There are two other characteristic modes at about 63.3 mHz (peak 2) and 216 mHz (peak 3).

FIG. 7. (Color online) Typical temporal changes in $\Delta \theta_7$, $\Delta \theta_8$, and $\Delta \theta_9$ observed for a certain neighboring pair of particles. When $N = 8$ and 9, the time period in which a certain mode exists becomes short.

FIG. 8. (Color online) Temporal changes in $\Delta \theta_6$ for particles in the $N = 6$ system starting from three specific configurations: (a) one doublet and four singlets, (b) two doublets and two singlets, and (c) three doublets. (a) and (b) display oscillatory motion. (c) displays steady configuration motion.
is due to the nonuniformity of the optical driving force. The lifetime of configuration A is shorter than that of the other configurations, and configuration A soon changes into another configuration. This agrees with the low stability of singlets in our system, as discussed above. The motion mainly observed in Fig. 6(a) is that of configurations B and C. The characteristic time period observed in Fig. 8(a) is about 12 s, and that in Fig. 8(b) is about 25 s. These values are almost in agreement with the periods corresponding to the peaks 1 and 2 in the frequency spectrum in Fig. 6(b). The switching between the relatively stable modes B and C is due to the fluctuation of the driving force, as shown in Fig. 6(a).

The stable collective modes in the same system have been studied theoretically by linear stability analysis of the equally spaced configuration [15]. A constant tangential driving force $F^\theta$ is applied to particles of radius $a$. The particles are constrained to a circular path of radius $R$ by a harmonic optical potential with force constant $K'$. The velocity of particle $i$, $d\mathbf{r}_i/dt$, is

$$\frac{d\mathbf{r}_i}{dt} = \mu_i^{\theta} F_i + \sum_{j \neq i} \mu_{ij}^{\theta} F_j,$$

where $\mathbf{r}_i$ is the position of particle $i$ relative to the center of the circle, $\mu_i^{\theta}$ is the mobility tensor, and $F_i$ is the force applied to particle $i$. $F_j = -K'(r_j - R)e_j + F^\theta e^\theta$. The unit vectors $e^\theta$ and $e^\theta$ are respectively parallel and perpendicular to $r_j$. By applying the Oseen approximation, $\mu_i^{\theta}$ is given as

$$\mu_{ij}^{\theta} = \frac{3\mu a}{4r_{ij}} \left( 1 + \frac{r_j \otimes r_{ij}}{r_{ij}^3} \right),$$

where $\mu = (6\pi \eta a)^{-1}$ is the mobility of a single particle in a medium of viscosity $\eta$, $\mu_{ij}^{\theta} = \mu^I (1$ is the unit tensor), $r_{ij} = r_i - r_j$, and $r_{ij} = |r_{ij}|$. The equation of motion for particle $i$ in the polar direction is

$$\frac{d\theta_i}{dt} = \mu^{\theta} F^\theta + \frac{3\mu a}{4} \sum_{j \neq i} \frac{1}{r_{ij}} \left[ F^\theta \left( \cos \theta_j + \frac{r_j r_{ij}}{r_{ij}^3} \sin^2 \theta_j \right) \right.$n\left. - K'(r_j - R) \left( 1 + \frac{r_j}{r_{ij}} (r_j - r_i \cos \theta_j) \right) \sin \theta_j \right],$$

where $\delta \theta_j = \theta_j - \theta_i$, $r_i = |r_i|$, and $r_j = |r_j|$. The stability of the equally spaced configuration with $\theta_i(t) = 2\pi i/N + \Omega N t$ in the case of a small radial deviation $\delta r_i$ and a small angular deviation $\delta \theta_i$ has been discussed by solving the linearized equations of motion given by Eq. (1) [15]. The angular velocity of an equally spaced cluster $\Omega_N$ is given as

$$\Omega_N = \frac{\mu^{\theta} F^\theta}{R} \left[ 1 + \frac{3}{4} \sum_{j \neq i} \frac{1 + 3 \cos \Phi_{ij}}{2X_{ij}} \right],$$

where $X_{ij} = \sqrt{2R/a} |1 - \cos \Phi_{ij}|$ and $\Phi_{ij} = 2\pi (i - j)/N$. In the following discussion, the force constant of the harmonic potential $K'$ is assumed to be very large, and only the angular fluctuation $\delta \theta_i$ is taken into account. Although the induction time required to start oscillatory behavior is infinitely long under this condition, its characteristic frequency can be obtained with precision of order $a/R$ even under the above assumption. The finally obtained eigenvalues in reduced units are

$$\lambda \delta \theta_i + \frac{3}{4} \sum_{j \neq i} \left[ \frac{1}{X_{ij}} \left( \frac{7 - 3 \cos \Phi_{ij}}{4(1 - \cos \Phi_{ij})} \right) \right] = 0,$$

where $\lambda$ is the complex eigenvalue and is scaled by $\mu^{\theta} F^\theta / R$.

When $N = 3$ and 4, there is a single oscillatory mode, $\lambda_3 = 17\sqrt{3}/32(a/R)$ and $\lambda_4 = 21\sqrt{3}/16(a/R), respectively. When N = 5, there are two oscillatory modes, $\lambda_{5(1)} = 3\sqrt{5}/8(a/R)$ and $\lambda_{5(2)} = 117\sqrt{2}/64(a/R). When N = 6, there are two oscillatory modes, $\lambda_{6(1)} = (2667 + 1683\sqrt{3}/2)^{1/2}/16(a/R)$ and $\lambda_{6(2)} = (2667 - 1683\sqrt{3}/2)^{1/2}/16(a/R). When N = 7, there are three oscillatory modes, but they cannot be expressed in a simple analytical form. The characteristic frequencies obtained by solving the linear equations given by Eq. (5) for $a/R = 1.5/10$ are plotted in Fig. 9. To compare these results with the experimental data, the theoretical values are converted into actual values using the angular velocity of a single particle of $\mu^{\theta} F^\theta / R = 1.0 \text{ rad/s}$. The characteristic frequencies of the oscillatory modes marked in the experimental spectra [Figs. 3(b), 4(b), 5(b), and 6(b)] are also shown in Fig. 9. For $N = 3–5$, both frequencies are in good agreement. For $N = 6$ and 7, the lowest frequencies exhibit reasonably good agreement, but the higher frequencies are markedly different. Since the stability of these faster modes is low, we cannot obtain sufficient temporal data to evaluate their characteristic frequencies from their experimental spectra.

### VI. EFFECT OF STABLE COLLECTIVE MOTION ON AVERAGE PARTICLE VELOCITY

Finally, we discuss the effect of the characteristic collective motion of the particles on their average motion by considering the mean angular velocity $\omega_N$ in systems with various values of $N$. Since the temporal fluctuation of $\theta_i$ originating from the reorganization of doublets is very small, the observed temporal evolution of the angular distance $\theta_i(t)$ almost linearly increases with the elapsed time $t$. The mean velocity $\omega_N$ simply calculated from the slope of $\theta_i(t)$ is shown in Fig. 10. The
velocity $\omega_N$ generally increases with $N$. This tendency can be explained by the reduced effective viscous drag of the particles. However, we found an odd-even effect for the systems with $N = 1$–7. A similar phenomena has also recently been observed up to $N = 11$ in an experimental study [22]. The steady configuration motion composed of stable and fast doublets was only observed in even-$N$ systems. Therefore, even-$N$ systems have lower viscous drag, resulting in their larger angular velocity than that expected from systems with neighboring values of $N$.

The dependence of $\omega_N$ on $N$ has also been studied theoretically [15]. Our experimental results are qualitatively in agreement with the theoretical results in terms of the tendency that $\omega_N$ increases with $N$. However, our results for small $N$ exhibit an “odd-even effect,” which is not predicted by the simulation. We presume that this effect originates from the existence of relatively stable steady configuration motion. The nonuniformity of the optical force is also not considered in the simulation. However, it is important to understand the mechanism underlying our experimental results because such nonthermal noise on the dynamics of our system is currently being studied by artificially modifying the optical path, and the results will be reported in the near future.

Although we only consider a single path in this study, one can make the experimental model for the collective motion in a carpet of microfluidic rotors [24] and a model of flagellar movement by extending the system to multiple paths. The collective motion reported in this paper may also indicate how the efficient transport of many colloidal particles of micrometer scales can be achieved and may offer some strategies for designing microfluidic devices.

VII. CONCLUSION

In the present paper, we have studied the collective motion of various numbers ($N = 2$–9) of optically driven particles on a circular path. Doublet clusters are spontaneously formed by the hydrodynamic interaction between the particles, and the doublets move faster than a singlet. This induces characteristic oscillatory and steady configuration motion in the angular difference between neighboring particles. With increasing $N$, various types of characteristic motion appear, and frequent switching between them is observed. This causes the dynamics of the system to become more complex in large-$N$ systems. An increase in $N$ also enhances the formation of transient larger clusters and increases the average velocity of the particles.

The oscillatory modes observed in the experiment are discussed theoretically by linear stability analysis and experimentally by setting the particles in a specific initial arrangement. The number of these modes is determined by the number of doublets appearing in the system. We can confirm the direct connection between a specific configuration of the particles and a specific behavior in the angular difference.

The origin of the observed switching between the stable types of motion is an important problem to be solved, which we leave as a future problem at this stage. There are many experimental factors that are related to such behavior. One can control the relative magnitude of the optical driving force, the hydrodynamic interaction, and the nonthermal noise by tuning the experimental conditions in our system. The effect of nonthermal noise on the dynamics of our system is currently being studied by artificially modifying the optical path, and the results will be reported in the near future.

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