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Collective motion of symmetric camphor papers in an annular water channel

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We investigate the collective motion of symmetric self-propelled objects that are driven by a difference in the surface tension. The objects move around an annular water channel spontaneously and interact through the camphor layer that develops on the water surface. We found that two collective motion modes, discrete and continuous density waves, are generated depending on the number of self-propelled objects. The two modes are characterized by examining the local and global dynamics, and the collective motion mechanism is discussed in relation to the distribution of camphor concentration in the annular water channel. We conclude that the difference between these two modes originates from that of the driving mechanism that pushes a camphor paper away from a cluster, through which mechanism density waves are generated and maintained.

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I. INTRODUCTION

Studies on autonomous motors that mimic their biological counterparts can assist not only in the understand of energy transduction and chemotaxis mechanisms in biological systems but also in the development of autonomous motors that can be used in medical and engineering fields for inspection, manipulation, and transportation tasks [1–3]. Several artificial systems that exhibit self-propelled motion have been studied under conditions close to isothermal and chemical nonequilibrium conditions [3–11], but it is difficult for the motion in these systems to change in response to the environment, such as achieving taxis in a flagellar motor.

We have previously investigated the characteristic features of self-propelled systems composed of camphor [11–13] for various internal conditions (e.g., scraping morphology [14,15] and the chemical structure of the camphor derivatives [16]) and external conditions (e.g., temperature, surface tension, chemical stimuli, and the shape of the cell [17,18]). Camphor systems are ideal for clarifying the mechanisms of collective motion in organisms since their experimental parameters are controllable as a simple inanimate system. The collective motion of asymmetric camphor boats in an annular water channel has also been reported [19–21]. Although theoretical research has suggested that symmetric and asymmetric self-propelled motors may have different motion features [22], the characteristic features of the collective motion of symmetric self-propelled objects still need to be realized experimentally.

We investigated the collective motion of symmetric self-propelled circular papers containing camphor in an annular water channel. We found that two collective motion modes, i.e., discrete and continuous density waves, are generated depending on the number of self-propelled papers. Especially, a discrete density wave has not been observed in the asymmetric motors.

II. EXPERIMENT

Methanol and (+)-camphor were purchased from Nacalai Tesque, Inc. (Kyoto, Japan) and Wako Pure Chemicals (Kyoto, Japan), respectively. The circular papers (10 mm diameter) were fabricated from the filter paper (WHATMAN, 5307-090, USA) using a brass punch and soaked in a saturated solution of camphor in methanol (1.1 g/ml) for several seconds [9]. These filter papers were then dried in air to remove the methanol (hereafter, referred to as "camphor paper"). To prepare the annular water channel, 11 ml of distilled water was poured onto a ring-shaped polyester film (inner diameter: 50 mm; outer diameter: 90 mm) placed on a Teflon plate. Since the hydrophobicity of the Teflon plate was larger than that of the polyester film, the water droplet formed an annular shape, as shown in Fig. 1. The average height of the water phase was 2.5 mm. The motion of the floating camphor papers was monitored with a digital video camera (HDR-CX560V, Sony, Tokyo, Japan; minimum time resolution: 1/30 s) in an air-conditioned room at 298 ± 2 K and then analyzed using an image-processing system (ImageJ, National Institutes of Health, MD, USA).

III. EXPERIMENTAL RESULTS

The characteristic feature of collective motion of the camphor papers changes depending on the number of camphor papers N_c. Figure 2 shows snapshots of the collective motion for $N_c = 1$, 7, and 14. When $N_c = 1$ [Fig. 2(a)], the camphor paper exhibits uniform motion in the channel. As the camphor paper is symmetric, the direction of motion depends on the initial condition. When more camphor papers are set, the motion changes from uniform to intermittent oscillatory motion (alternating between rapid and slow motion). In the latter case, while most of the papers exhibited slow motion (without the white arrow in Fig. 2), a density wave propagated locally around the channel. Under these conditions, we found that two collective motion modes were generated depending on N_c. When $N_c = 7$ [Fig. 2(b)], each camphor paper began to move after being pushed by a paper behind it, subsequently stopping after colliding with the preceding camphor paper. The same processes continuously propagated along the channel. We call this type of collective motion "mode I." On the other hand, when $N_c = 14$ [Fig. 2(c)], each camphor paper began to move after the preceding paper had moved away; the camphor paper then caught up with the preceding stationary paper. We call this type of collective motion "mode II." See the movie in the Supplemental Material [23].

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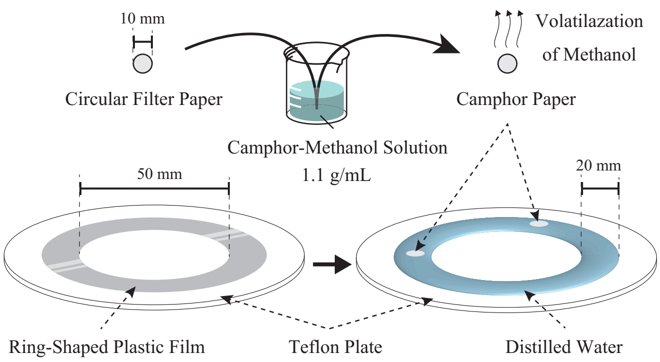

FIG. 1. (Color online) Experimental setup. Filter paper cut into 10-mm-diam circles was soaked in a saturated solution of camphor in methanol for several seconds. The dried filter papers are referred to as "camphor papers." To form the annular channel, 11 ml of distilled water was poured onto a ring-shaped plastic film placed on a Teflon plate.

To qualitatively evaluate the differences in the collective motion from the local dynamics, we introduce the parameter $\theta_j^{\pm}$ defined as follows:

$$\theta_j^{\pm} = |\theta_{j\pm1} - \theta_j|, \tag{1}$$

where θ_j ($j = 1 - N_c$) is the central angle of the jth camphor paper (P_j) relative to the annular channel, as shown in Fig. 3(a). If $\partial\theta_j^{\pm}/\partial t > 0$, the distance between P_j and $P_{j\pm1}$ increases, while it decreases if $\partial\theta_j^{\pm}/\partial t < 0$. For $N_c = 1$, 7, and 14, time-space (t-θ_j) diagrams are shown in the upper part of Fig. 3(b), and the lower figures show the relationship between θ^+ and θ^-, which corresponds to the bold lines in the upper figures. The trajectories were independent of papers. The individual configurations of the camphor papers around the jth paper were maintained for a few seconds when the conditions corresponded to those in a corner of the triangular orbit where $\theta^+ \approx \theta^-$. In fact, the camphor papers were balanced at a characteristic separation angle for a short time. We call this position the "balanced state," i.e., the camphor papers exhibit slow motion to maintain this state. In the case of mode I, the characteristic angle was close to the average angle, $2\pi/(N_c - 1)$, while in the case of mode II, the characteristic angle was close that when the papers were in contact.

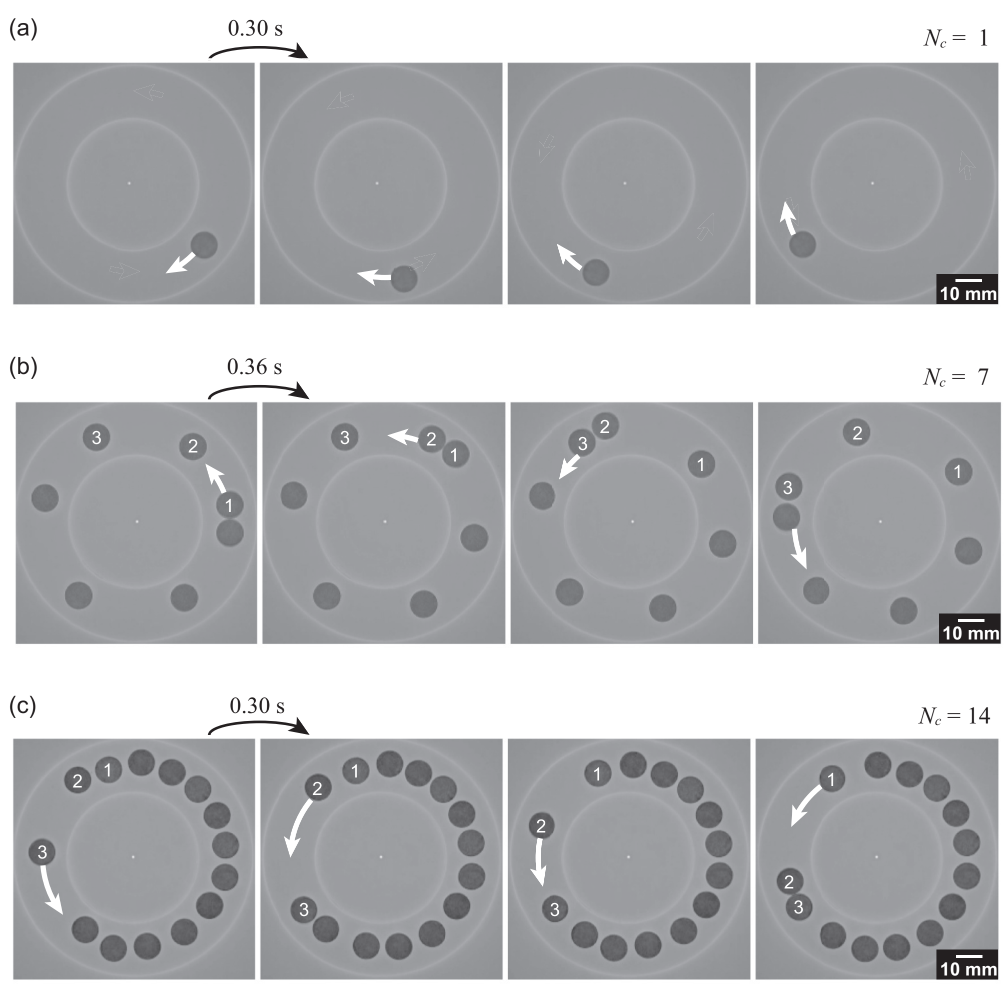

FIG. 2. Snapshots of the collective motion for $N_c =$ (a) 1, (b) 7, and (c) 14 (top view). The time intervals of the snapshots were (a) 0.30, (b) 0.36, and (c) 0.30 s. The white arrow denotes the direction of motion of the camphor paper. The papers without a white arrow do not move, or hardly move. The number ($i = 1$, 2, or 3) on the camphor paper indicates the different camphor papers (P_i).

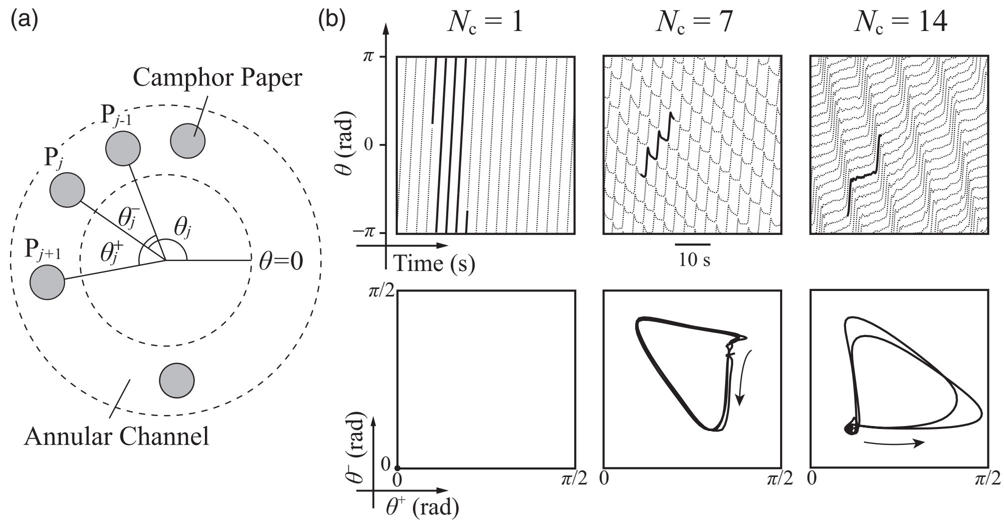

FIG. 3. Different collective motion from the local point of view. (a) θ_j ($j = 1 - N_c$) is the central angle of the camphor paper (P_j) relative to the annular channel. (b) The upper figures show time-space (t-θ_j) diagrams, and the lower figures show the relationship between θ^+ and θ^-, corresponding to the bold lines in the upper figures, for $N_c = 1$, 7, and 14.

To quantitatively evaluate the dependence of the number of camphor papers on the collective motion from a global dynamics, we introduce an order parameter δ_{N_c} as follows:

$$\delta_{N_c} = \left| \frac{1}{N_c} \sum_{k=1}^{N_c} e^{i\theta_k} \right|. \tag{2}$$

If the camphor papers are located at equal intervals in the annular water channel, then $\delta_{N_c} = 0$. Moreover, the maximum value of δ_{N_c} ($\delta_{N_c,\,\max}$) corresponds to the configuration in which all the camphor papers are in contact, i.e., the camphor papers are maximally crowded. This $\delta_{N_c,\,\max}$ value is given by

$$\delta_{N_c,\,\max} = \left| \frac{1}{N_c} \sum_{k=1}^{N_c} e^{ik\theta_{\min}} \right|, \tag{3}$$

where $\theta_{\min}$ is the angle between two touching papers, as shown in Fig. 4(a). As $\delta_{N_c,\,\max}$ depends on N_c, we introduced a normarized order parameter Δ as follows:

$$\Delta = \frac{\delta_{N_c}}{\delta_{N_c,\,\max}}. \tag{4}$$

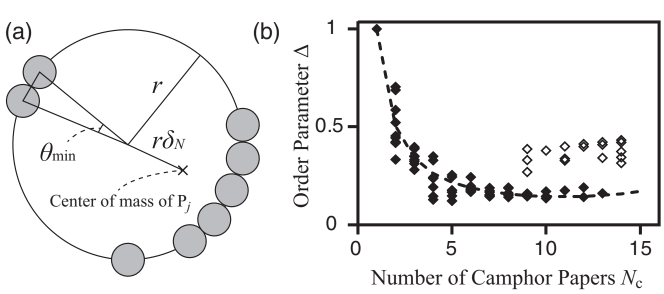

FIG. 4. Dependence of the number of camphor papers on collective motion for global dynamics. (a) $\theta_{\min}$ is the angle between two papers in contact, and $r\delta_{N_c}$ is the center mass of P_j. (b) The relationship between the number of camphor papers N_c and the normalized order parameter Δ. The diamonds show the average of Δ over 100 s for each experiment. The open diamonds indicate that mode II is the dominant mode of the collective motion.

The larger Δ is, the more is the configuration bias of camphor papers. Figure 4(b) shows the dependence of the collective motion on the number of camphor papers. The dashed line is defined by $\Delta(n)$ ($n \in \mathbf{R}$):

$$\Delta(n) = \frac{1}{n\delta_{n,\,\max}}. \tag{5}$$

This relation was obtained by assuming that the configuration of $n - 1$ stationary camphor papers constitutes a regular polygon while then another one moves. The diamonds show the average Δ value over 100 s for each experiment. For each N_c, at least four experiments were performed. According to Fig. 3(b), if mode I is dominant, then Δ takes a value close to the dashed line since the camphor papers, which are almost stationary, are located at the characteristic angle $2\pi/(N_c - 1)$, i.e., a regular polygon is formed by the camphor papers [solid diamonds in Fig. 4(b)]. If mode II is dominant, then Δ takes a value larger than that given by Eq. (5), as the camphor papers are almost crowded [open diamonds in Fig. 4(b)]. With an increase in N_c, the collective motion of the camphor papers changes from mode I to mode II, and there is an interval in which both modes are observed. During this interval, the collective motion occasionally transited between both modes. The fluctuations of the open diamonds in Fig. 4(b) are owing to the transition between both modes.

IV. DISCUSSION

Based on the present experimental results and previous work on the self-motion of camphor objects [13,20–22], the camphor papers are propelled around an annular water channel by the difference in the surface tension induced by the camphor molecule itself. Since the driving force is balanced at a viscous

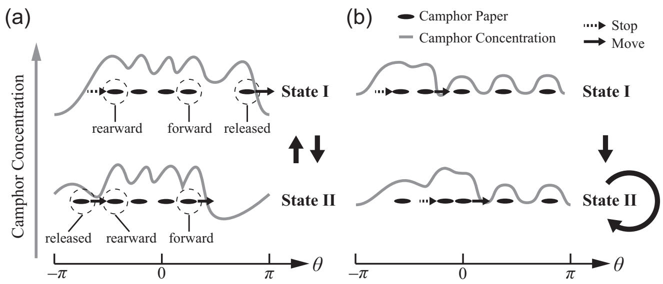

FIG. 5. Schematic illustration of the mechanism for (a) mode II and (b) mode I. The camphor papers are represented by the black ellipses and the gray line shows the camphor concentration on the water surface.

resistance, the camphor paper moves uniformly, as shown in Fig. 2(a) [13].

Figure 3(b) suggests that the collective motion in mode I [Fig. 2(b)] is qualitatively different from that in mode II [Fig. 2(c)]. That is, the difference in the angle ($\theta^{\pm}$) between neighboring camphor papers decreases and increases in a different order from the balanced state in modes I and II, respectively. Considering the mechanism of mode II first, Fig. 4(b) suggests that it is easy to maintain a cluster of camphor papers as N_c is increased. One camphor paper (referred to as the "released paper") is released from the edge of a cluster and then joins the same cluster at another end (the "rearward paper"). The camphor paper that was located immediately behind the released paper (the "forward paper") cannot move due to the influence of the camphor from the released paper [state I in Fig. 5(a)]. In addition, the rearward paper cannot move in the reverse direction due to the influence of its own camphor. Furthermore, the intervening camphor papers hardly move since there is a low repulsive force in an area with a high camphor concentration [19–22]. The forward paper is released from the cluster when the camphor from the previously released paper is sublimated [state II in Fig. 5(a)], i.e., the forward paper is released when the forward surface tension increases. The released paper joins the cluster before the rearward paper begins to move due to sublimation of its own camphor. In this case, motion is realized by alternating between states I and II, and the cluster in density wave propagates around the channel to time.

To explain the mechanism of mode I, we assume that the camphor papers are uniformly distributed around the channel due to an equivalent repulsive force. From this configuration, we suppose that one paper is driven by a fluctuation in the camphor concentration [state I in Fig. 5(b)]. When this paper approaches the neighboring paper, it forms a two-paper cluster. The forward paper is then released since the rearward surface tension has decreased, and the rearward paper is momentarily stationary owing to the influence of its own camphor and that of the released paper. The original two-paper cluster disappears [state II in Fig. 5(b)] but another is formed by the released paper. This appearance and disappearance of the two-paper clusters are indicated by the relationship between θ^+ and θ^- for $N_c = 7$ in Fig. 3(b), i.e., either θ^+ or θ^- is immediately turned up near $\theta_{\rm min}$. The motion of the camphor papers is realized by iterating state II. That is, a discrete density wave propagates around the annular water channel. In both modes, one camphor paper begins to move by increasing the relative surface tension in front of the paper. While the rearward surface tension decreases in mode I, the forward surface tension increases in mode II.

In this way, the dynamical feature of the system remarkably changes depending on the number of camphor papers. As imagined easily, it depends also on the number density and the size of the camphor papers, and the dominant factor among them varies according to the situation of the system. We reported that the collective motion depends on the number density of the camphor boat [20]. If the size of the camphor paper is small, the region of mode II in Fig. 4(b) may shift to the higher N_c since the amount of camphor molecules supplied from the small camphor paper to the water surface is low. The effect of the water volume and the number of camphor paper will be reported separately by evaluating the effect of collision of camphor paper and the boundary effect of the chamber in a future work.

V. CONCLUSIONS

Our study has revealed that two collective motion modes of symmetric self-propelled objects are generated depending on whether the angular difference between neighboring objects increases or decreases from the balanced state. The collective motion mechanisms were discussed in terms of the camphor concentration on the water surface. We concluded that the time variation of the angular difference depends on how the motion of the camphor paper is initiated. An increase in the forward surface tension or a decrease in the rearward surface tension will initiate the mode of motion and determine the collective motion characteristics.

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[23] See Supplemental Material at http://link.aps.org/supplemental/10.1103/PhysRevE.88.012911 for the movie of the two collective motion modes.