Rotational motion of a camphor disk in a circular region

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In a two-dimensional axisymmetric systems, a symmetric self-propelled particle exhibits rotational or oscillatory motion, from the consideration of the system symmetry. In this paper, we studied motion of a camphor disk confined in a two-dimensional circular region. By reducing the model describing the dynamics of the motion of a camphor disk and the concentration field of camphor molecules on water surface, we analyzed the bifurcation structure where the rest state at the center of the system becomes unstable. As a result, we found that rotational motion is stably realized through the double-Hopf bifurcation from the rest state. The theoretical results were confirmed by numerical calculation and well corresponded to the experimental results.

I. INTRODUCTION

There are a lot of systems which show self-propelled motion [1–4]. In self-propelled systems, particles move consuming and dissipating free energy. In such systems, the symmetric property is one of the classificatory criteria. First of all, self-propelled systems are classified into symmetric and asymmetric systems. A symmetric self-propelled particle moves through spontaneous symmetry breaking, while one with asymmetry moves in the direction predetermined by the asymmetry. Here we focus on a symmetric self-propelled system.

In a two-dimensional axisymmetric system, rotational and oscillatory motion are often observed [2,10]. Bacteria confined in a quasi-two dimensional circular chamber exhibit rotational motion. A laser-heated oil droplet exhibits oscillatory and rotational motions depending on the laser intensity [6]. Phenomenological models which can reproduce rotational or oscillatory motion have been proposed [4,11]. To understand which motion, rotation or oscillation, is realized in a two-dimensional axisymmetric system, we have discussed criteria by a theoretical approach based on a weakly nonlinear analysis [12]. In the analysis, only two assumptions were imposed: One is that a self-propelled particle is confined by a harmonic potential. The other is that the system is near the bifurcation point where rest state at the bottom of the harmonic potential becomes unstable. However, our proposed criteria have not been applied to actual systems. A camphor-water system is the one of the candidate to which our criteria can be applied.

The camphor-water system is one of the physico-chemical self-propelled systems [13,14]. When a camphor particle is placed onto water surface, it releases camphor molecules onto the water surface. The released camphor molecules reduce the surface tension. In other words, camphor particle releases its repellants (camphor molecules) and moves to the region with less repellents. The camphor-water system is so simple that various geometries can be realized [18–27]. For example, the shape and size of water chamber can be one of the parameters which can affect the motion of the camphor particle [18,19]. The shape of the particle is also can be changed, e.g., the motion of an elliptic camphor particle [21,22] and the interaction of them [23] have been investigated. The mathematical model reflecting the elementary processes is also available [17,28,29]. The model consists of two equations: one is the equation of motion describing the dynamics of the position of the camphor particle, and the other is the reaction-diffusion equation describing the concentration field of camphor molecules at water surface. From the viewpoint of the theoretical analysis, the model for the camphor-water systems is easy to analyze since the reaction-diffusion equation for the concentration field of camphor molecules on water surface has a reaction term described by a piecewise linear function. The advantage of the camphor-water system is that the theoretical and experimental approaches are both available [17,18,24,25]. In addition, a camphor-water system is considered to be one of the simplest negative-chemotactic systems [11], and can be applied in other systems [30,51].

In this paper, we consider the dynamics of a camphor particle confined in a two-dimensional circular water chamber as a good example of a symmetric self-propelled particle in a two-dimensional axisymmetric system, and apply our previously proposed criteria to it in order to analyze which motion, rotation or oscillation, is selected. First, we introduce the mathematical model constructed based on the elemental phenomena. Then we reduce it and apply the result of the weakly nonlinear analysis. The theoretical result has been compared with the numerical and experimental results.

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is expressed as:

\[ F(\rho(t)) = \lim_{\epsilon \to +0} \frac{-k}{\pi \epsilon^2} \int_0^{2\pi} [c(\rho) + \epsilon n(\theta) \cdot \nabla c(\rho)] \epsilon d\theta \]

\[ = -k \nabla c|_{r=\rho} \cdot . \]

Here, we assume that the surface tension \( \gamma \) is a linear decreasing function with regard to the camphor concentration \( c \), i.e., \( \gamma = -kc + \gamma_0 \), where \( k \) and \( \gamma_0 \) are positive constants. \( \gamma_0 \) gives the surface tension of pure water. Hereafter, we consider the following equation as for the motion of a camphor disk:

\[ \sigma \frac{d^2 \rho}{dt^2} = -\xi \frac{d\rho}{dt} + F(\rho, \rho), \]

The time evolution for concentration field \( c = c(r, t) \) is described by the following equation:

\[ \frac{\partial c}{\partial t} = D \left( \frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} + \frac{1}{r^2} \frac{\partial^2}{\partial \theta^2} \right) c - \alpha c + f, \]

where \( r = (r, \theta) \) in polar coordinates is an arbitrary position in the circular region, \( D \) is the effective diffusion constant, \( \alpha \) is the sublimation rate, and \( f \) denotes the dissolution of camphor molecules from the camphor disk. The source term \( f \) is given by

\[ f(r, \rho) = \frac{c_0 \alpha}{\pi \epsilon^2} \Theta(\epsilon - |r - \rho|) = \begin{cases} \frac{c_0 \alpha}{\pi \epsilon^2}, & (|r - \rho| < \epsilon), \\ 0, & (|r - \rho| > \epsilon), \end{cases} \]

where \( \Theta(\cdot) \) denotes the Heaviside step function and \( c_0 \) is the total amount of the dissolution from the camphor disk per unit time. By taking the limit that \( \epsilon \) goes to +0, the source term is expressed as

\[ f(r, \rho) = \frac{c_0 \alpha}{\pi \epsilon^2} \delta(r - \rho) \delta(\theta - \phi), \]

\[ \delta(\cdot) \] denotes the Dirac’s delta function and \( c_0 \) is the dissolution per unit time. Here we assume that the camphor molecules dissolve constantly at the position of the camphor disk \( \rho = (\rho(t), \phi(t)) \) in polar coordinates. The Neumann boundary condition:

\[ \frac{\partial c}{\partial r} \bigg|_{r=R} = 0, \]

is imposed to Eq. (9).

In the following analysis, dimensionless forms of Eqs. (2), (3), (4), (5), and (6),

\[ \sigma \frac{d^2 \rho}{dt^2} = -\xi \frac{d\rho}{dt} + \tilde{F}(\tilde{c}; \tilde{\rho}), \]
\[ \mathbf{F}(\rho, \mathbf{i}) = \lim_{\epsilon \rightarrow +0} \frac{1}{S} \int_{\partial \Omega} \tilde{\gamma}(c(\rho + \epsilon n)) \mathbf{n} \cdot d\mathbf{r} \]  
\[ = - \tilde{\nabla} \tilde{c} \bigg|_{\mathbf{r} = \tilde{\rho}}. \]  
(11)

\[ \frac{\partial \tilde{c}}{\partial t} = \left( \frac{\partial^2}{\partial \mathbf{r}^2} + \frac{1}{\mathbf{r}} \frac{\partial}{\partial \mathbf{r}} + \frac{1}{\mathbf{r}^2} \frac{\partial^2}{\partial \theta^2} \right) \tilde{c} - \tilde{c} + \tilde{f}, \]  
(13)

\[ \tilde{f}(\mathbf{r}; \tilde{\rho}) = \delta(\mathbf{r} - \tilde{\rho}), \]  
(14)

\[ \frac{\partial \tilde{c}}{\partial \mathbf{r}} \bigg|_{\mathbf{r} = \tilde{\mathbf{r}}} = 0, \]  
(15)

are used. The details of nondimensionalization are shown in Appendix \[ A \]. Hereafter, we omit tilde (\( \tilde{\} \)).

It should be noted that the hydrodynamic effect is neglected in the present model. In this system, the inhomogeneities of surface tension is brought by camphor concentration gradient and then the Marangoni flow occurs \[ [32, 33] \]. The profile of the concentration field of camphor molecules become wider due to the Marangoni flow, which can be included as the effective diffusion constant in Eq. \[ 10 \] \[ [34] \].

B. Reduction of the Model

The equations are reduced around the rest state where the camphor disk stops at the center position of the circular region. First, the concentration field \( c \) is expanded with Bessel function and Fourier series for radial and angular directions, respectively, as

\[ c(r, \theta, t) = \frac{1}{2\pi} \sum_{m=-\infty}^{\infty} \sum_{n=0}^{\infty} a_{nm} c_{mn}(t) J_m(k_{mn} r) e^{i m \theta}, \]  
(16)

where \( J_m \) is the first-kind Bessel function of \( m \)-th order. Here \( a_{nm} = -2/(R^2 J_m^m(\zeta_n) J_m(\zeta_n)) \) is a normalization constant, where \( J_m(\zeta_n) \) is the \( n \)-th local extrema, i.e., \( J_m(\zeta_i) = J_m(\zeta_j) = 0 \), and \( \zeta_i < \zeta_j \) for \( i < j \). The details of the expansion are shown in Appendix \[ B \]. The source term in Eq. \[ 10 \] is also expanded as

\[ f(r, \theta; \rho, \phi) = \frac{1}{2\pi} \sum_{m=-\infty}^{\infty} \sum_{n=0}^{\infty} a_{nm} J_m(k_{mn} \rho(t)) \times J_m(k_{mn} r) e^{i m (\theta - \phi(t))}. \]  
(17)

Thus we have the equation for concentration in wavenumber space:

\[ \frac{d c_{mn}}{dt} = -(k_{mn}^2 + 1) c_{mn} + J_m(k_{mn} \rho(t)) e^{-im \phi(t)}. \]  
(18)

First, the Green function \( g_{mn}(t) \) is calculated. The Green function is defined as a function that satisfies the following equation:

\[ \frac{dg_{mn}}{dt} = -(k_{mn}^2 + 1) g_{mn} + \delta(t). \]  
(19)

By solving the above equation, we have

\[ g_{mn}(t) = \begin{cases} e^{-(k_{mn}^2 + 1)t} & (t > 0) \\ 0 & (t < 0) \end{cases} = e^{-(k_{mn}^2 + 1)t} \Theta(t), \]  
(20)

where \( \Theta(t) \) is the Heaviside step function.

Using the Green’s function \( g_{mn} \), the concentration field \( c_{mn} \) in wavenumber space is described as

\[ c_{mn}(\rho) = \int_{-\infty}^{t} J_m(k_{mn} \rho(t')) e^{-im \phi(t')} e^{-(k_{mn}^2 + 1)(t-t')} dt'. \]  
(21)

By expanding the above integration with regard to the time using partial integration, we have the concentration field in the wavenumber space expanded with regard to the current position, velocity, acceleration, and so on \[ [13, 24, 35] \]. Then the expanded concentration field is converted into the real space. The detailed calculations are presented in Appendix \[ B \]. Using the definition of the driving force in Eq. \[ 11 \], the driving force \( \mathbf{F} \) is expressed as follows:

\[ \mathbf{F}(\rho, \dot{\rho}, \ddot{\rho}) = a(R) \rho + b(R) \dot{\rho} + c(R) |\rho|^2 \rho + g(R) \ddot{\rho} \]  
\[ + b(R) |\dot{\rho}|^2 \rho + j(R) (\rho \cdot \dot{\rho}) \rho + k(R) |\rho|^2 \dot{\rho} \]  
\[ + n(R) |\rho|^2 \ddot{\rho} + p(R) (\rho \cdot \dot{\rho}) \ddot{\rho}, \]  
(22)

where \( \mathbf{F} \) is the function of \( \rho, \dot{\rho}, \) and \( \ddot{\rho} \). The coefficients of the terms in Eq. \[ 22 \] are explicitly obtained as:

\[ a(R) = \frac{1}{4\pi} \left( \frac{K_0'(R)}{T_0(R)} + \frac{K_1'(R)}{T_1(R)} \right), \]  
(23)

\[ b(R) = \frac{1}{4\pi} \left( -\gamma_{\text{Euler}} + \log \frac{2}{e} \right) \]  
\[ + \frac{1}{8\pi} \left( 2 \frac{K_1'(R)}{T_1(R)} + \left( 1 + \frac{1}{R^2} \right) \frac{1}{(T_1(R))^2} \right), \]  
(24)

\[ c(R) = \frac{1}{32\pi} \left( 3 \frac{K_0'(R)}{T_0(R)} + 4 \frac{K_1'(R)}{T_1(R)} + \frac{K_2'(R)}{T_2(R)} \right), \]  
(25)

\[ g(R) = -\frac{1}{16\pi} \left( 1 + \left( R + \frac{1}{R} \right) \frac{T_1''(R)}{(T_1(R))^3} - \frac{1}{(T_1(R))^2} \right), \]  
(26)
We have the following dynamical system by the reduction:

\[ h(R) = \frac{1}{64\pi} \left( \frac{8K'_0(R)}{I'_0(R)} + \frac{K'_1(R)}{I'_1(R)} - \frac{4K'_2(R)}{I'_2(R)} - 2R \frac{I''_0(R)}{(I'_0(R))^3} - (R + \frac{1}{R}) \frac{I''_1(R)}{(I'_1(R))^3} + \frac{2}{R^2} + 3 \right) \frac{1}{(I'_1(R))^2} - \frac{1}{8(R^2 + 3)} \frac{1}{(I'_2(R))^2}, \]

\[ j(R) = \frac{1}{16\pi} \left( \frac{4K'_0(R)}{I'_0(R)} + \frac{4K'_1(R)}{I'_1(R)} + \frac{1}{(I'_0(R))^2} + \left( \frac{1}{1 + \frac{1}{R^2}} \right) \frac{1}{(I'_1(R))^2} \right), \]

\[ k(R) = \frac{1}{128\pi} \left( -4 + 3(1 + R^2) \frac{I''_0(R)}{(I'_1(R))^2} + \frac{4}{(I'_0(R))^2} \right) \frac{I''_0(R)}{(I'_1(R))^3} - \frac{3}{R + 7R} \frac{I''_0(R)}{(I'_1(R))^3} - (1 + R^2) \frac{I''_1(R)}{(I'_1(R))^3}, \]

\[ n(R) = \frac{1}{32\pi} \left( \frac{1}{1 + \frac{1}{R^2}} \right) \frac{1}{(I'_1(R))^2} + \left( \frac{4}{R^2} + 3 \right) \frac{1}{(I'_2(R))^2}, \]

\[ p(R) = \frac{1}{32\pi} \left( 4 \frac{K'_1(R)}{I'_1(R)} + 4 \frac{K'_2(R)}{I'_2(R)} - \frac{1}{R + 1} \frac{I''_2(R)}{(I'_1(R))^3} - \frac{4}{R^2 + 3} \frac{I''_1(R)}{(I'_1(R))^3} \right) + \left( \frac{8}{R^2 + 3} \right) \frac{1}{(I'_2(R))^2}, \]

where \( \gamma_{\text{Euler}} \) is the Euler-Mascheroni constant \( (\gamma_{\text{Euler}} \approx 0.577). \)

We confirmed that, when \( R \) goes to infinity, \( a(R), c(R), h(R), j(R), n(R), \) and \( p(R) \) go to zero, and \( b(R), g(R), \) and \( k(R) \) go to \(-\gamma_{\text{Euler}} + \log(2/e)/(4\pi)\), \(-1/(16\pi)\), and \(-1/(32\pi)\), respectively. Such dependences of coefficients on \( R \) are consistent with the result for the case where a camphor disk is located in an infinite two-dimensional region \[24].

### C. Bifurcation Analysis

We have the following dynamical system by the reduction of the proposed model for a camphor disk confined in a circular region as:

\[ (\sigma - g(R))\dot{\rho} = a(R)\rho + (b(R) - \xi)\dot{\rho} + c(R)\rho^2\rho + h(R)|\dot{\rho}|^2\rho + j(R)(\rho \cdot \dot{\rho}) + k(R)|\dot{\rho}|^2\rho + n(R)|\rho|^2\dot{\rho} + p(R)(\rho \cdot \dot{\rho})\dot{\rho}, \]

In this dynamical system, double-Hopf bifurcation occurs at \( b(R) = \xi \), where the coefficient of \( \dot{\rho} \) is zero. When \( \xi \) is greater than \( b(R) \), the rest state where the camphor particle stops at the center position of the circular region is stable. On the other hand, the rest state becomes unstable. The profile of \( b(R) \) has a single peak as shown in Fig. 2, which is similar to the case of the one-dimensional system \[18].

To clarify what motion occurs in the case that the rest state is unstable, we apply the results obtained by weakly nonlinear analysis reported in our previous paper \[12\]. Here we assume that the terms, \((\sigma - g(R))\dot{\rho}\) and \(a(R)\rho\), which cause harmonic oscillation, are the main terms, and the other terms are the perturbative terms. We convert time \( t \) to \( \tau = \omega t \), where \( \omega(R, \sigma) = \sqrt{-a(R)/(\sigma - g(R))} \), which is the rescaling of the time with regard to the frequency of the harmonic oscillation. Then we have

\[ \dot{\rho} = -\rho + (B(R, \sigma) - \Xi)\dot{\rho} + C(R, \sigma)|\rho|^2\rho + H(R)|\dot{\rho}|^2\rho + J(R, \sigma)(\rho \cdot \dot{\rho}) + K(R, \sigma)|\dot{\rho}|^2\rho + N(R, \sigma)|\rho|^2\dot{\rho} + P(R)(\rho \cdot \dot{\rho})\dot{\rho}, \]

where \( B(R, \sigma) = b(R)/\omega(R, \sigma), \Xi = \xi/\omega(R, \sigma), C(R, \sigma) = c(R)/\omega(R, \sigma)^2, H(R) = h(R), J(R, \sigma) = j(R)/\omega(R, \sigma), K(R, \sigma) = k(R)\omega(R, \sigma), N(R, \sigma) = n(R)/\omega(R, \sigma), \) and \( P(R) = p(R). \)

In our previous paper \[12\], we have derived the conditions for stable rotation:

\[ \begin{cases} K(R, \sigma) + N(R, \sigma) < 0, \\ K(R, \sigma) - N(R, \sigma) + J(R, \sigma) < 0, \end{cases} \]

and for stable oscillation:

\[ \begin{cases} 3K(R, \sigma) + N(R, \sigma) + J(R, \sigma) < 0, \\ K(R, \sigma) - N(R, \sigma) + J(R, \sigma) > 0. \end{cases} \]
FIG. 3: Plots of $F_{\text{osc}}(R) = K(R, \sigma) + N(R, \sigma)$, $F_{\text{rot}}(R) = 3K(R, \sigma) + N(R, \sigma) + J(R, \sigma)$, and $F_{\text{crt}}(R) = K(R, \sigma) - N(R, \sigma)$ against the radius of the circular region $R$, where $\sigma$ is set to be 0.01. Rotational motion is linearly stable in a certain range of $R$, which is indicated by coloring with magenta.

By applying the above criteria to Eq. (33), it is concluded that the rotational motion is stably observed when the radius of the circular region is around the diffusion length ($\sim 1$), as shown in Fig. 3.

III. NUMERICAL CALCULATION

To confirm the theoretical results, we performed numerical calculations based on Eq. (10) with the driving force:

$$\tilde{F}(\tilde{\rho}(t)) = \frac{1}{S} \int_{\partial \tilde{\Omega}} \tilde{\gamma}(\tilde{\ell}(\tilde{\rho} + \epsilon m)) \cdot \tilde{n} \cdot d\tilde{r},$$

and Eq. (13) with the source term:

$$f(r; \rho) = \frac{1}{\pi \epsilon^2} \Theta (\epsilon - |r - \rho|).$$

Here the size of the camphor disk is considered to be finite ($\epsilon = 0.1$) in order to avoid difficulty originating from the treatment of the Dirac's delta function in numerical calculation. We used the Euler method for Eq. (10) and the explicit method for Eq. (13). The time and spatial steps were $10^{-5}$ and $10^{-2}$, respectively. The mass per unit area $\sigma$ was fixed to $\sigma = 10^{-2}$. In order to calculate the force acting on the camphor disk in Eq. (36), we adopted the summation over 40 arc elements in the place of the integration in Eq. (11).

Here we show the numerical results for the radius of the circular region $R = 1$. The results for the resistance coefficient per unit area $\xi = 0.18$ and $\xi = 0.2$ are shown in Figs. 4 and 5 respectively. The initial conditions were the same. We obtained the trajectories toward the circular orbit whose center corresponds to the center of the circular region for $\xi = 0.18$ and toward the rest state at the center of the circular region for $\xi = 0.2$. Thus it is expected that the bifurcation point exists between $\xi = 0.18$ and $\xi = 0.2$. The bifurcation point for the radius of the circular region $R = 1$ expected by the theoretical analysis is ca. 0.218. The order of the bifurcation point is the same as that obtained by the numerical results though there still remains some difference between them. We guess the dominant reason of the difference of theoretical and numerical results in the bifurcation point is the difference of the size of the camphor disk. In the theoretical analysis, the size of the camphor disk is assumed to be $\epsilon = +0$, but in the numerical calculation, $\epsilon$ has a finite value $\epsilon = 0.1$.

FIG. 4: Numerical results on the trajectories of a camphor disk in a circular region for the resistance coefficient $\xi = 0.18$. The camphor disk exhibited rotational motion. (a) The trajectory on the $x$-$y$ plane. (b) Time evolutions of $x(t)$ and $y(t)$ shown in blue- and red-colored curves, respectively. The initial conditions for the position and velocity of the camphor disk were $x = 0.1, y = 0.2, v_x = -0.01$, and $v_y = 0$, respectively. The initial concentration field $c$ was zero at every point in the region.
IV. EXPERIMENTS

We also made experiments to confirm the theoretical results. Here we observed motion of a camphor disk in the circular water chamber whose radius was continuously controlled.

A. Experimental setup

A camphor gel disk, whose diameter was 4.0 mm and thickness was 0.5 mm, was made of agar gel in which water was replaced with camphor methanol solution. After the methanol dried up, a camphor disk was floated on a water phase (15 mm in the depth). To achieve a variable-sized water phase, an optical focus (IDC-025, Sigma-koki) was placed on the water phase whose radius \( R \) could be changed. As an initial state, a camphor disk was placed on a water chamber with a small radius (\( R = 5.0 \) mm) where the disk was in the rest state. Then, the radius was increased to 13.0 mm and the motion of the camphor disk was monitored. The details of the experimental setup are shown in Appendix E.

B. Experimental results

At the initial stage with the small radius of water phase (\( R \sim 5.0 \) mm), the disk was in the rest state. With an increase in the radius of the circular water chamber \( R \), the disk started to move and finally showed rotational motion as shown in Fig. 6(a). For rotational motion, both the moving speed \( v \) and the position of the disk \( r \) were almost constant in time as shown in Fig. 6(b). The theoretical results qualitatively explain the transition from the rest state at the center position of the circular water chamber to the rotational motion with an increase in the radius of the circular water chamber \( R \). At first, the disk transiently showed reciprocal motion, and then, transited to rotational motion (Figure 1b). The duration time of reciprocal motion tended to be shorter in a larger radius of the circular water chamber.

V. DISCUSSION

The mathematical model for motion of a camphor disk in a two-dimensional circular region was reduced into the ordinary differential equation of the second order. In the reduction of the concentration field, the discrete Hankel transform with Neumann condition is applied to the reaction-diffusion equation in Eq. (13), the modified Helmholtz equation. The discrete Hankel transform with Neumann condition is applied to the Helmholtz equation as in Eq. (5.3.34) in Ref. [38]. Then the bifurcation structure of the reduced equation is analyzed. By changing the radius of the circular region or the resistance coefficient, the rest state at the center position of the circular region becomes unstable through the double-Hopf bifurcation \([37]\). When the radius of the circular region is around the diffusion length, the double-Hopf bifurcation is supercritical and rotational motion of a camphor disk with an infinitesimally small radius is stably observed. For larger radius of the circular region, we guess that double-Hopf bifurcation might be subcritical, since similar dependence on the radius of the circular region is seen in the case of one-dimensional finite region. The reason which determines whether the double-Hopf bifurcation is supercritical or subcritical has not been clearly revealed. However, it is expected that the branch of the radius of rotational motion rises perpendicularly to the parameter axis at the bifurcation when \( R \) goes infinity since the transrational motion emerges when the rest state becomes unstable in an infinite region.
The bifurcation type also depends on the mass density of the camphor disk. The line which shows the boundary between the parameter regions where the double-Hopf bifurcation is supercritical or not is shown in Fig. 7.

When the rest state becomes unstable, the camphor disk moves toward the boundary in a certain direction to avoid the region with higher concentration of camphor molecules, which were seen in both numerical calculations and experiments. Then the camphor particle is reflected by interacting with the boundary through the concentration field, and moves toward the other side with regard to the center of the circular region. Thus the camphor disk first exhibits a reciprocal motion as shown in Figs. 4 and 5. Since the oscillation is unstable, the reciprocal motion is transient and a rotational motion is finally realized.

VI. CONCLUSION

In the present paper, we discussed motion of a camphor disk confined in a two-dimensional circular water chamber. We reduced the mathematical model for the camphor disk motion, and applied our previous results based on the weakly nonlinear analysis to the reduced equation. The theoretical results suggest that the rotational motion occurs when the rest state becomes unstable for a water chamber whose radius is comparable with the order of the diffusion length. The theoretical results were confirmed by numerical calculation and experiments.

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[1] S. Ramaswamy, Annu. Rev. Cond. Mat. Phys. 1, 323 (2010).
[2] T. Ohta, J. Phys. Soc. Jpn. 86, 072001 (2017).
Appendix A: Dimensionless form of the mathematical model

We consider the nondimensionalization of Eq. 6. The dimensions of $\alpha$, $D$, and $c_0$ are $[1/T]$, $[L^2/T]$, and $[C/L^2]$, respectively. Here, $T$, $L$, and $C$ represent the dimensions of time, length, and concentration, respectively. Thus, we introduce the dimensionless time, position, and concentration as $\tilde{t} = at$, $\tilde{r} = \sqrt{\alpha/D} r$, and $\tilde{c} = c/c_0$, respectively. By substituting the three dimensionless variables into Eq. 6 and dividing the both sides of the above equation with $c_0$, we obtain

$$
\frac{\partial \tilde{c} (\tilde{r}, \tilde{\theta}, \tilde{\phi})}{\partial \tilde{t}} = \left( \frac{\partial^2}{\partial \tilde{r}^2} + \frac{1}{\tilde{r}} \frac{\partial}{\partial \tilde{r}} + \frac{1}{\tilde{r}^2} \frac{\partial^2}{\partial \tilde{\theta}^2} \right) \tilde{c} (\tilde{r}, \tilde{\theta}, \tilde{\phi}) - \tilde{c} (\tilde{r}, \tilde{\theta}, \tilde{\phi}) + \frac{1}{c_0 \alpha} f \left( \sqrt{\frac{D}{\alpha}} \tilde{r}, \tilde{\theta}; \sqrt{\frac{D}{\alpha}} \tilde{\rho} (\tilde{t}), \tilde{\phi} (\tilde{t}) \right).
$$

(A1)

The source term is considered as follows:

$$
\frac{1}{c_0 \alpha} f \left( \sqrt{\frac{D}{\alpha}} \tilde{r}, \tilde{\theta}; \sqrt{\frac{D}{\alpha}} \tilde{\rho} (\tilde{t}), \tilde{\phi} (\tilde{t}) \right) = \frac{1}{\tilde{r}} \delta (\tilde{r} - \tilde{\rho} (\tilde{t})) \delta (\tilde{\theta} - \phi (\tilde{t})) \equiv \tilde{f} (\tilde{r}, \tilde{\theta}; \tilde{\rho}, \tilde{\phi}).
$$

(A2)

Here we use $\delta (ax) = \delta (x)/|a|$. Then, we have

$$
\frac{\partial \tilde{c} (\tilde{r}, \tilde{\theta}, \tilde{\phi})}{\partial \tilde{t}} = \left( \frac{\partial^2}{\partial \tilde{r}^2} + \frac{1}{\tilde{r}} \frac{\partial}{\partial \tilde{r}} + \frac{1}{\tilde{r}^2} \frac{\partial^2}{\partial \tilde{\theta}^2} \right) \tilde{c} (\tilde{r}, \tilde{\theta}, \tilde{\phi}) - \tilde{c} (\tilde{r}, \tilde{\theta}, \tilde{\phi}) + \tilde{f} (\tilde{r}, \tilde{\theta}; \tilde{\rho}, \tilde{\phi}),
$$

(A3)

where $\tilde{\rho} = \sqrt{\alpha/D} \rho$.

Next, Eq. 3 is nondimensionalized. The variables $t$, $r$, $\rho$, and $c$ are replaced with $\tilde{t}$, $\tilde{r}$, $\tilde{\rho}$, and $\tilde{c}$, and then we have

$$
\sigma D \frac{d^2 \tilde{\rho} (\tilde{t})}{d\tilde{t}^2} = -\xi \sqrt{\alpha} D \frac{d \tilde{\rho} (\tilde{t})}{d\tilde{t}} + F \left( \frac{c_0}{\alpha}, \tilde{c} \right).
$$

(A4)
In Eq. (A3), we cannot eliminate all coefficients but one. Here, we adopt the dimensionless driving force,

$$\tilde{F}(\rho; \tilde{c}) = \frac{c_0}{\sqrt{\alpha D}} \times$$

$$\lim_{\tilde{c} \to \infty} - \frac{\Gamma}{\pi \epsilon^2} \int_0^{\infty} \tilde{c} \left( \sqrt{\frac{1}{\epsilon \rho}} (\tilde{\rho} + \tilde{m}(\theta)) : \sqrt{\frac{1}{\epsilon \alpha}} (\rho) \right) \tilde{c} d\theta$$

$$= \frac{c_0 \Gamma}{\sqrt{\alpha D}} \tilde{F}(\tilde{\rho}; \tilde{c}).$$  \hspace{1cm} (A5)

Here, $\tilde{F}$ is a dimensionless driving force. Then we obtain

$$\frac{\sigma \alpha^2 D d^2 \tilde{\rho}}{\Gamma c_0} \frac{dt^2}{dt} = - \frac{\xi \alpha D d \tilde{\rho}}{\Gamma c_0} + \tilde{F}(\tilde{\rho}; \tilde{c}),$$  \hspace{1cm} (A6)

where

$$\tilde{\sigma} \equiv \frac{\sigma \alpha^2 D}{\Gamma c_0}, \quad \tilde{\xi} \equiv \frac{\xi \alpha D}{\Gamma c_0}.$$  \hspace{1cm} (A7)

Thus dimensionless forms in Eqs. (10) to (15) are obtained.

Appendix B: Discrete Hankel transform

Here, we provide some notes on “discrete Hankel transform” for a function which satisfies the Neumann condition [36, 38, 39]. In the book by Bowman [36], the discrete Hankel transform for a function which satisfies the Dirichlet condition is denoted, and as for the case with Neumann boundary condition, the calculation can be proceeded in almost a parallel manner.

The Bessel differential equation is given as

$$J_n''(\lambda) + \lambda J_n'(\lambda) + \lambda_2 \lambda J_n(\lambda) = 0.$$  \hspace{1cm} (A8)

By replacing $r$ with $\lambda r$, we have

$$\left( \frac{d^2}{dr^2} + \frac{1}{r} \frac{d}{dr} + \left( \lambda^2 - \frac{\nu^2}{r^2} \right) \right) J_n(\lambda r) = 0.$$  \hspace{1cm} (B2)

Equation (B2) is transformed in the following form:

$$\frac{d}{dr} \left( r \frac{d J_n(\lambda r)}{d r} \right) - \frac{\nu^2}{r} J_n(\lambda r) + \lambda^2 r J_n(\lambda r) = 0.$$  \hspace{1cm} (B3)

Here we consider the difference between equations in Eq. (B3) with different $\lambda$. We have

$$\frac{J_n'(\lambda n r) d}{dr} \left( r \frac{d J_n(\lambda m r)}{d r} \right) - J_n(\lambda m r) \frac{d}{dr} \left( r \frac{d J_n(\lambda n r)}{d r} \right)$$

$$+ (\lambda_n^2 - \lambda_m^2) r J_n(\lambda m r) J_n(\lambda n r) = 0.$$  \hspace{1cm} (B4)

Using Eq. (B4), we have

$$- (\lambda_n^2 - \lambda_m^2) \int_0^R r J_n(\lambda m r) J_n(\lambda n r) dr$$

$$= \left[ J_n(\lambda n r) \left( r \frac{d J_n(\lambda m r)}{d r} \right) - J_n(\lambda m r) \left( r \frac{d J_n(\lambda n r)}{d r} \right) \right]_0^R.$$  \hspace{1cm} (B5)

Here we set $\lambda_n \equiv \zeta_n / R$, where $J_n(\zeta_n)$ is the $n$-th local extrema, i.e., $J_n(\zeta_i) = J_n(\zeta_j) = 0$, and $\zeta_i < \zeta_j$ for $i < j$. Since \((\partial J_n(\lambda m r)) / (\partial r) = 0\) holds considering the Neumann condition, we have

$$\int_0^R (\lambda_n^2 - \lambda_m^2) r J_n(\lambda m r) J_n(\lambda n r) dr = 0,$$  \hspace{1cm} (B6)

For $m \neq n$, we have

$$\int_0^R r J_n(\lambda m r) J_n(\lambda n r) dr,$$  \hspace{1cm} (B7)

and thus $J_n(\lambda m r)$ and $J_n(\lambda n r)$ whose domains are $[0, R]$ are orthogonal to each other for $m \neq n$.

To obtain the norm of $J_n(\lambda n r)$, we calculate the following integration using Eq. (B5):

$$\int_0^R r J_n(\lambda n r) J_n(\lambda n r) dr$$

$$= \lim_{\lambda \to \lambda_n} \int_0^R r J_n(\lambda n r) J_n(\lambda n r) dr$$

$$= \lim_{\lambda \to \lambda_n} R \left( \lambda J_n(\lambda n R) J_n'(\lambda n R) - \lambda_n J_n(\lambda n R) J_n'(\lambda n R) \right) / \lambda^2 - \lambda_n^2.$$  \hspace{1cm} (B8)

By applying L’Hôpital’s rule, we have

$$\int_0^R r J_n(\lambda n r) J_n(\lambda n r) dr$$

$$= \frac{R^2}{2} \left( J_n'(\lambda n R) J_n(\lambda n R) \right) - \left( \frac{1}{\lambda_n^2} J_n'(\lambda n R) J_n(\lambda n R) \right) J_n(\lambda n R).$$  \hspace{1cm} (B9)

Since $J_n'(\lambda n R) = 0$ holds considering the Neumann condition, we have

$$\int_0^R r J_n(\lambda n r) J_n(\lambda n r) dr$$

$$= \frac{R^2}{2} J_n''(\lambda n R) J_n(\lambda n R) = \frac{R^2}{2} J_n''(\zeta_n) J_n(\zeta_n) \equiv \frac{1}{a_{\nu n}}.$$  \hspace{1cm} (B10)

Thus the functions $\{\sqrt{a_{\nu n}} J_n(\lambda n r)\}$ are the basis of the function space for $[0, R]$. A function $f(r)$ which satisfies the Neumann condition at $r = R$ is expressed in the “discrete Hankel transform” as

$$f(r) = \sum_{n \in \mathbb{N}} a_{\nu n} f_n \lambda_n J_n(\lambda n r),$$  \hspace{1cm} (B11)

where

$$f_n \equiv \int_0^R f(r) J_n(\lambda n r) r dr.$$  \hspace{1cm} (B12)

Appendix C: Reduction of the mathematical model

Equation (21) is expanded using the partial integration as follows [33]:
The explicit form of the steady state is obtained as
\[ c(r, \rho(t)) = \frac{1}{2\pi} \sum_{m=-\infty}^{\infty} \sum_{n=0}^{\infty} \frac{a_{m|n}}{k_{m|n}^2 + 1} J_{|m|}(k_{m|n}\rho(t))J_{|m|}(k_{m|n}r)e^{im(\theta - \phi(t))} \]

only depends on the current position of the camphor disk, and corresponds to the steady state concentration field when the camphor disk is fixed at the current position for long time. The explicit form of the steady state is obtained as
\[ c_0(r, \theta, \rho(t), \phi(t)) = \frac{1}{2\pi} K_0 \left( \sqrt{r^2 + \rho^2 - 2r\rho \cos(\theta - \phi)} \right) \]

The detailed calculation is presented in Appendix D. First, the length of Eqs. (C2) and (C3) is rescaled with \( \lambda \) as \( r = \lambda r, \rho = \lambda \rho R = \lambda R, k_{m|n} = k_{m|n}/\lambda \). Then the both sides of the rescaled Eqs. (C2) and (C3) are differentiated with regard to \( \lambda, \rho, \) and \( \phi \). By setting \( \lambda = 1 \), the expressions with modified Bessel functions for the other terms in Eq. (C1) are obtained.
the driving force is calculated as

\[ \text{in the circular region is obtained. The steady state in an} \]

\[ \text{tion field for a fixed camphor disk at an arbitrary position} \]

\[ \text{by taking the terms up to the third-order of} \ \rho \ \text{and} \ \dot{\rho}, \]

\[ \text{C4} \]

By taking the terms up to the third-order of \( \rho \) and \( \dot{\rho} \), and first-order of \( \ddot{\rho} \), the driving force as a function of the trajectory of the camphor disk and the radius of the circular region is obtained as in Eq. (22).

\[ \text{Appendix D: Derivation of the steady concentration field} \]

In this section, the derivation of the steady concentration field for a fixed camphor disk at an arbitrary position in the circular region is obtained. The steady state in an infinite region is obtained as follows:

\[ c(r, \theta) = \frac{1}{2\pi} K_0 \left( \sqrt{r^2 + \rho^2 - 2r\rho \cos(\theta - \phi)} \right). \]
The detailed derivation is referred in Ref. [24]. To satisfy the Neumann boundary condition, we adequately add the general solution for Eq. (13) without the source term, i.e., the homogeneous form of Eq. (13):

\[
\left( \frac{\partial^2}{\partial r^2} + \frac{1}{r} \frac{\partial}{\partial r} + \frac{1}{r^2} \frac{\partial^2}{\partial \theta^2} \right) g(r, \theta) - g(r, \theta) = 0.
\]  

(D2)

From the definition of the modified Bessel functions, the general solution of Eq. (D2) is expressed as

\[
c(r, \theta) = A_0 K_0(r) + B_0 I_0(r) \\
+ \sum_{m=1}^{\infty} \left( A_m K_m(r) + B_m I_m(r) \right) \cos m(\theta - \phi) \\
+ \sum_{m=1}^{\infty} \left( C_m K_m(r) + D_m I_m(r) \right) \sin m(\theta - \phi).
\]

(D3)

By considering the symmetric property of the system, the \(m\)-th mode term should be expressed only by \(\cos m(\theta - \phi)\), i.e., \(C_m\) and \(D_m\) should be zero. Furthermore, \(K_n(r)\) \((n \geq 1)\) is not suitable for representing the concentration field of camphor, since the integration,

\[
\int_0^{2\pi} \int_0^R K_n(r) r dr d\theta,
\]

(D4)
diverges for \(n \geq 1\). \(K_0(r)\) diverges at \(r = 0\) and is not suitable when a camphor disk is off the origin. When a camphor disk is located at the origin, \(K_0(r)\) is already included as the steady state without the Neumann boundary. Thus, for the both cases, the concentration field with the correction terms should be given by the following form:

\[
c(r, \theta) = \frac{1}{2\pi} B_0 \left( \sqrt{r^2 + \rho^2 - 2\rho r \cos(\theta - \phi)} \right)
\]

\[
+ \sum_{m=0}^{\infty} B_m I_m(r) \cos m(\theta - \phi).
\]

(D5)

Then, the coefficients \(B_m\) are determined by the boundary condition in Eq. (13), that is

\[
\left. \frac{1}{2\pi} \frac{\partial}{\partial r} K_0 \left( \sqrt{r^2 + \rho^2 - 2\rho r \cos(\theta - \phi)} \right) \right|_{r=R} = -\sum_{m=0}^{\infty} B_m \left. \frac{\partial I_m(r)}{\partial r} \cos m(\theta - \phi) \right|_{r=R}.
\]

(D6)

If \(\partial K_0 \left( \sqrt{r^2 + \rho^2 - 2\rho r \cos(\theta - \phi)} \right) / \partial r\) at \(r = R\) is expanded with regard to \(\cos m(\theta - \phi)\), we can determine \(B_m\). By using the formula for \(R > r\) which is represented in Eq. (8) in p.361 of Ref. [40]:

\[
K_0 \left( \sqrt{R^2 + r^2 - 2Rr \cos \theta} \right) = \sum_{n=-\infty}^{\infty} K_n(r) I_n(r) \cos n\theta,
\]

(D7)

we have

\[
\frac{\partial}{\partial R} \int_0^{2\pi} K_0 \left( \sqrt{R^2 + \rho^2 - 2\rho \rho \cos(\theta - \phi)} \right) \cos n(\theta - \phi) d\theta
\]

\[
= 2\pi \frac{\partial K_n(R)}{\partial R} I_n(\rho) \quad (n = 0, 1, 2, \cdots).
\]

(D8)

Here we use \(K_{-m}(r) = K_m(r)\) and \(I_{-m}(r) = I_m(r)\). As a consequence, we have

\[
B_0 = \frac{1}{2\pi} \frac{\partial I_0(R)}{\partial R} I_0(\rho),
\]

(D9)

\[
B_n = \frac{1}{\pi} \frac{\partial I_n(R)}{\partial R} \frac{\partial I_n(\rho)}{\partial \rho} \quad (n = 1, 2, \cdots)
\]

(D10)

Thus we have Eq. (13). It should be noted that the conservation of integration of concentration over the circular region,

\[
\int_0^R \int_0^{2\pi} c(r, \theta) r dr d\theta = 1,
\]

(D11)

is satisfied.

**Appendix E: Detail Experimental Conditions**

Mili-Q water (360 mL) was poured into a plastic container (150 mm in width, 150 mm in length, and 30 mm in depth) and the optical focus (Sigma-koki, IDC-025) was placed on the water surface with silicone sheet as a spacer (15 mm in the depth). The optical focus has a circular hole whose radius \(R\) was able to change from 0.5 to 20.0 mm. To prepare the camphor disk, agar gel sheet was replaced with camphor MeOH solution, the gel sheet was soaked into methanol (MeOH) at first, and then, cut into circular shape. A camphor MeOH solution (1.0 g/mL) for more than 12 hours. After the MeOH in an agar gel was completely rinsed with water and was cut into circular shape. A camphor gel disk was floated on the water phase and its moving behavior was observed with a video camera (handycam, Sony, video rate: 30 fps). The radius of the water phase \(R\) was about 5.0 mm at first, and then, increased to 13.0 mm or 16.1 mm with a constant speed (4.0 mm s\(^{-1}\)).