

Rotational motion of traveling spots in dissipative systems

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What is the origin of rotational motion? An answer is presented through the study of the dynamics for spatially localized spots near codimension 2 singularity consisting of drift and peanut instabilities. The drift instability causes a head-tail asymmetry in spot shape, and the peanut one implies a deformation from circular to peanut shape. Rotational motion of spots can be produced by combining these instabilities in a class of three-component reaction-diffusion systems. Partial differential equations dynamics can be reduced to a finite-dimensional one by projecting it to slow modes. Such a reduction clarifies the bifurcational origin of rotational motion of traveling spots in two dimensions in close analogy to the normal form of 1:2 mode interactions.

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Spatially localized moving objects such as traveling pulses and self-propelled particles are fundamental objects arising in nonlinear science, which display a large variety of dynamical behaviors in many dissipative systems [1–4]. In two dimensions, traveling motion causes symmetry breaking from the circular shape of a standing spot. The onset of a straight motion of traveling spots has been studied for a drift instability [3,5,6], in which the authors showed a drift pitchfork scenario from the local properties at a codimension 1 bifurcation. This topic has attracted much interest experimentally as well as theoretically with respect to driven droplet patterns on a solid substrate [7–9]. In biological tissues, digital image analysis also shows that a head-tail asymmetry in cell shape determines the direction of motion and some sorts of interference wave pattern occurs during spontaneous cell migration [10,11]. These recent experiments allow us to deduce the underlying mechanism of interplay between the spot locomotion and shape-change dynamics.

In this paper, we consider the spot dynamics near a codimension 2 singularity for reaction-diffusion systems in which the associated parameter values are located close to the drift and peanut bifurcation points. Drift instability originates in the translation-free mode and the associated deformation eigenvector represents a \mathcal{D}_1 symmetry breaking from a disk shape. Peanut one is by \mathcal{D}_2 symmetry-breaking bifurcation, corresponding to two-mode deformation, where \mathcal{D}_n stands for the dihedral symmetry group. We show that such a codimension 2 singularity can induce rotational motion of traveling spots—that is, rotational spot (RS) motion—in a class of reaction-diffusion systems.

The occurrence of such a motion is generic because the original partial differential equations (PDEs) can be reduced to finite-dimensional ordinary differential equations (ODEs) based on the center manifold theory [12–14], and the resulting ODEs take a normal form of 1:2 mode interaction of cubic type. We analyze the reduced ODEs, and show that there exists a solution in which both drift velocity vector and peanut deformation become time-periodic functions that correspond to the rotational motion of traveling spot solution to the original reaction-diffusion systems. The information specific to the form and parameters of the original PDEs is con-

tained in the coefficients of the reduced system. We also discuss about the relationship between the global bifurcational structures of the original PDEs and the reduced ODEs, which sheds light on the origin of rotational motion, that is, such a motion emerges through the interaction between drift and peanut instabilities and it is realized in the PDE counterpart, i.e., the three-component reaction-diffusion system.

A general setup for the PDE system in a neighborhood of codimension 2 bifurcation point $\lambda^c = (\lambda_1^c, \lambda_2^c)$ reads, with a small parameter $\eta = (\eta_1, \eta_2)$ as $\lambda = \lambda^c + \eta$,

$$\mathbf{u}_t = D\Delta\mathbf{u} + F(\mathbf{u}; \lambda) \equiv \mathcal{L}(\mathbf{u}; \lambda^c) + \sum_{i=1}^2 \eta_i \mathbf{g}_i(\mathbf{u}), \quad (1)$$

where \mathbf{g}_i ($i=1, 2$) is N -dimensional vector-valued functions. Let $X := \{L^2(\mathbb{R})\}^N$, $\mathbf{u}(t, \mathbf{r}) = (u_1, \dots, u_N)^T \in X$, be an N -dimensional vector and $F: \mathbb{R}^N \rightarrow \mathbb{R}^N$, D be a positive diagonal matrix. We assume that the nontrivial standing spot solution $S(\mathbf{r}; \lambda)$ exists at $\lambda = \lambda^c$, i.e., $\mathcal{L}(S; \lambda^c) = 0$.

Let L be the linearized operator $L = \mathcal{L}'[S(\mathbf{r}; \lambda^c)]$. L has a codimension 2 singularity at $\lambda = \lambda^c$ consisting of drift and peanut bifurcations in addition to the translation-free 0 eigenvalue; that is, there exist three types of eigenfunctions $\phi_i(\mathbf{r})$, $\psi_i(\mathbf{r})$, and $\xi_i(\mathbf{r})$ ($i=1, 2$) such that $L\phi_i = 0$, $L\psi_i = -\phi_i$, and $L\xi_i = 0$, where $\phi_i = \partial S / \partial x_i$ and ψ_i represents the deformation vector with Jordan form for the drift bifurcation. ξ_i^* is the \mathcal{D}_2 -symmetry breaking eigenfunction of peanut shape.

Similar properties also hold for L^* , that is, there exist ϕ_i^* , ψ_i^* , and ξ_i^* such that $L^*\phi_i^* = 0$, $L^*\psi_i^* = -\phi_i^*$, and $L^*\xi_i^* = 0$. Let $E = \text{span}\{\phi_i, \psi_i, \xi_i\}$ and the eigenfunctions be normalized by $\langle \psi_i, \phi_j \rangle_{L^2} = \langle \psi_i, \psi_j^* \rangle_{L^2} = 0$, and

$$\langle \phi_i, \psi_j^* \rangle_{L^2} = \langle \psi_i, \phi_j^* \rangle_{L^2} = \langle \xi_i, \xi_j^* \rangle_{L^2} = \begin{cases} \pi, & i = j \\ 0, & i \neq j. \end{cases} \quad (2)$$

The motion of a spot solution \mathbf{u} is essentially described by the two-dimensional vector functions of time t ; $\mathbf{p} = (p_1, p_2)$ denotes the location of the spot; $\mathbf{q} = (q_1, q_2)$ denotes its velocity; and $\mathbf{s} = (s_1, s_2)$ denotes its deformation. For small η , we can approximate a solution \mathbf{u} by

$$U = \tau(\mathbf{p}) \left\{ S(\mathbf{r}) + \sum_{i=1}^2 q_i \boldsymbol{\psi}_i(\mathbf{r}) + \sum_{i=1}^2 s_i \boldsymbol{\xi}_i(\mathbf{r}) + \boldsymbol{\zeta}^\dagger \right\}, \quad (3)$$

where $\tau(\mathbf{p})$ is the translation operator with $[\tau(\mathbf{p})\mathbf{u}](\mathbf{r}) = \mathbf{u}(\mathbf{r}-\mathbf{p})$. The remaining term $\boldsymbol{\zeta}^\dagger$ belongs to E^\perp . More precisely, $\boldsymbol{\zeta}^\dagger = q_1^2 \boldsymbol{\zeta}_1 + q_2^2 \boldsymbol{\zeta}_2 + q_1 q_2 \boldsymbol{\zeta}_3 + s_1^2 \boldsymbol{\zeta}_4 + s_2^2 \boldsymbol{\zeta}_5 + s_1 s_2 \boldsymbol{\zeta}_6 + q_1 s_1 \boldsymbol{\zeta}_7 + q_2 s_2 \boldsymbol{\zeta}_8 + q_1 s_2 \boldsymbol{\zeta}_9 + q_2 s_1 \boldsymbol{\zeta}_{10} + \eta_1 \boldsymbol{\zeta}_{11} + \eta_2 \boldsymbol{\zeta}_{12}$ with $\boldsymbol{\zeta}_k (k=1, \dots, 12) \in E^\perp$ are defined by solutions of

$$\begin{aligned} L\boldsymbol{\zeta}_1 + \frac{1}{2}F''(S)\boldsymbol{\psi}_1^2 + \boldsymbol{\psi}_{1x_1} &= \alpha\boldsymbol{\xi}_1, \\ L\boldsymbol{\zeta}_2 + \frac{1}{2}F''(S)\boldsymbol{\psi}_2^2 + \boldsymbol{\psi}_{2x_2} &= -\alpha\boldsymbol{\xi}_1, \\ L\boldsymbol{\zeta}_3 + F''(S)\boldsymbol{\psi}_1\boldsymbol{\psi}_2 + \boldsymbol{\psi}_{1x_2} + \boldsymbol{\psi}_{2x_1} &= 2\alpha\boldsymbol{\xi}_2, \end{aligned} \quad (4)$$

and

$$\begin{aligned} L\boldsymbol{\zeta}_4 + \frac{1}{2}F''(S)\boldsymbol{\xi}_1^2 &= 0, \\ L\boldsymbol{\zeta}_5 + \frac{1}{2}F''(S)\boldsymbol{\xi}_2^2 &= 0, \\ L\boldsymbol{\zeta}_6 + F''(S)\boldsymbol{\xi}_1\boldsymbol{\xi}_2 &= 0, \end{aligned} \quad (5)$$

and

$$\begin{aligned} L\boldsymbol{\zeta}_7 + F''(S)\boldsymbol{\psi}_1\boldsymbol{\xi}_1 + \boldsymbol{\xi}_{1x_1} &= \beta\boldsymbol{\psi}_1 + \beta'\boldsymbol{\phi}_1, \\ L\boldsymbol{\zeta}_8 + F''(S)\boldsymbol{\psi}_2\boldsymbol{\xi}_2 + \boldsymbol{\xi}_{2x_2} &= \beta\boldsymbol{\psi}_1 + \beta'\boldsymbol{\phi}_1, \\ L\boldsymbol{\zeta}_9 + F''(S)\boldsymbol{\psi}_1\boldsymbol{\xi}_2 + \boldsymbol{\xi}_{2x_1} &= \beta\boldsymbol{\psi}_2 + \beta'\boldsymbol{\phi}_2, \\ L\boldsymbol{\zeta}_{10} + F''(S)\boldsymbol{\psi}_2\boldsymbol{\xi}_1 + \boldsymbol{\xi}_{1x_2} &= -\beta\boldsymbol{\psi}_2 - \beta'\boldsymbol{\phi}_2, \end{aligned} \quad (6)$$

and

$$\begin{aligned} L\boldsymbol{\zeta}_{11} + \mathbf{g}_1(S) &= 0, \\ L\boldsymbol{\zeta}_{12} + \mathbf{g}_2(S) &= 0, \end{aligned} \quad (7)$$

where α , β , and β' are constants satisfying the following conditions:

$$\begin{aligned} \langle F''(S)\boldsymbol{\psi}_1\boldsymbol{\psi}_2 + \boldsymbol{\psi}_{1x_2} + \boldsymbol{\psi}_{2x_1} - 2\alpha\boldsymbol{\xi}_2, \boldsymbol{\xi}_2^* \rangle_{L^2} &= 0, \\ \langle F''(S)\boldsymbol{\psi}_1\boldsymbol{\xi}_2 + \boldsymbol{\xi}_{2x_1} - \beta\boldsymbol{\psi}_2 - \beta'\boldsymbol{\phi}_2, \boldsymbol{\phi}_2^* \rangle_{L^2} &= 0, \\ \langle F''(S)\boldsymbol{\psi}_1\boldsymbol{\xi}_2 + \boldsymbol{\xi}_{2x_1} - \beta\boldsymbol{\psi}_2 - \beta'\boldsymbol{\phi}_2, \boldsymbol{\psi}_2^* \rangle_{L^2} &= 0. \end{aligned} \quad (8)$$

Substituting Eq. (3) into Eq. (1) and taking the inner product with the adjoint eigenfunctions, the principal part of the reduced ODEs for (p_i, q_i, s_i) is given by

$$\dot{z}_0 = z_1 - \beta'\bar{z}_1 z_2,$$

$$\dot{z}_1 = M_1|z_1|^2 z_1 + M_2|z_2|^2 z_1 + M_3 z_1 + \beta\bar{z}_1 z_2,$$

$$\dot{z}_2 = N_1|z_2|^2 z_2 + N_2|z_1|^2 z_2 + N_3 z_2 + \alpha z_1^2. \quad (9)$$

Here we introduce the complex variables $z_0 = p_1 + ip_2$, $z_1 = q_1 + iq_2$, and $z_2 = s_1 + is_2$. Note that $\boldsymbol{\zeta}^\dagger$ with the conditions of Eqs. (4)–(7) is necessary for computations of cubic terms in Eq. (9).

The constants M_1 , M_2 , and M_3 are obtained from the model system (1) as follows:

$$\begin{aligned} \pi M_1 &= \frac{1}{6} \langle F'''(S)\boldsymbol{\psi}_1^3, \boldsymbol{\phi}_1^* \rangle_{L^2} + \langle F''(S)\boldsymbol{\psi}_1\boldsymbol{\zeta}_1, \boldsymbol{\phi}_1^* \rangle_{L^2} + \langle \boldsymbol{\zeta}_{1x_1}, \boldsymbol{\phi}_1^* \rangle_{L^2}, \\ \pi M_2 &= \frac{1}{2} \langle F'''(S)\boldsymbol{\xi}_1^2\boldsymbol{\psi}_1, \boldsymbol{\phi}_1^* \rangle_{L^2} + \langle F''(S)\boldsymbol{\psi}_1\boldsymbol{\zeta}_4, \boldsymbol{\phi}_1^* \rangle_{L^2} \\ &\quad + \langle F''(S)\boldsymbol{\xi}_1\boldsymbol{\zeta}_7, \boldsymbol{\phi}_1^* \rangle_{L^2} + \langle \boldsymbol{\zeta}_{4x_1}, \boldsymbol{\phi}_1^* \rangle_{L^2} - \beta' \langle \boldsymbol{\xi}_{1x_1}, \boldsymbol{\phi}_1^* \rangle_{L^2}, \\ \pi M_3 &= \eta_1 \langle F''(S)\boldsymbol{\psi}_1\boldsymbol{\zeta}_{11}, \boldsymbol{\phi}_1^* \rangle_{L^2} + \langle \mathbf{g}'_1(S)\boldsymbol{\psi}_1, \boldsymbol{\phi}_1^* \rangle_{L^2} \\ &\quad + \langle \boldsymbol{\zeta}_{11x_1}, \boldsymbol{\phi}_1^* \rangle_{L^2} + \eta_2 \langle F''(S)\boldsymbol{\psi}_1\boldsymbol{\zeta}_{12}, \boldsymbol{\phi}_1^* \rangle_{L^2} \\ &\quad + \langle \mathbf{g}'_2(S)\boldsymbol{\psi}_1, \boldsymbol{\phi}_1^* \rangle_{L^2} + \langle \boldsymbol{\zeta}_{12x_1}, \boldsymbol{\phi}_1^* \rangle_{L^2}. \end{aligned} \quad (10)$$

The constants N_1 , N_2 , and N_3 are also obtained as follows:

$$\pi N_1 = \frac{1}{6} \langle F'''(S)\boldsymbol{\xi}_1^3, \boldsymbol{\xi}_1^* \rangle_{L^2} + \langle F''(S)\boldsymbol{\xi}_1\boldsymbol{\zeta}_4, \boldsymbol{\xi}_1^* \rangle_{L^2},$$

$$\begin{aligned} \pi N_2 &= \frac{1}{2} \langle F'''(S)\boldsymbol{\psi}_1^2\boldsymbol{\xi}_1, \boldsymbol{\xi}_1^* \rangle_{L^2} + \langle F''(S)\boldsymbol{\psi}_1\boldsymbol{\zeta}_7, \boldsymbol{\xi}_1^* \rangle_{L^2} \\ &\quad + \langle F''(S)\boldsymbol{\xi}_1\boldsymbol{\zeta}_1, \boldsymbol{\xi}_1^* \rangle_{L^2} + \langle \boldsymbol{\zeta}_{7x_1}, \boldsymbol{\xi}_1^* \rangle_{L^2} - \beta' \langle \boldsymbol{\psi}_{1x_1}, \boldsymbol{\xi}_1^* \rangle_{L^2}, \end{aligned}$$

$$\begin{aligned} \pi N_3 &= \eta_1 \langle F''(S)\boldsymbol{\xi}_1\boldsymbol{\zeta}_{11}, \boldsymbol{\xi}_1^* \rangle_{L^2} + \langle \mathbf{g}'_1(S)\boldsymbol{\xi}_1, \boldsymbol{\xi}_1^* \rangle_{L^2} \\ &\quad + \eta_2 \langle F''(S)\boldsymbol{\xi}_1\boldsymbol{\zeta}_{12}, \boldsymbol{\xi}_1^* \rangle_{L^2} + \langle \mathbf{g}'_2(S)\boldsymbol{\xi}_1, \boldsymbol{\xi}_1^* \rangle_{L^2}. \end{aligned} \quad (11)$$

The bifurcation properties are determined by the coefficients of Eqs. (10) and (11), especially their signs, to the reduced systems. The derivations are shown in Appendixes A and B.

The dynamics of Eq. (9) are essentially governed by the last two equations, exactly the same as the normal form obtained in the study of resonance patterns in a bilayer fluid under $O(2)$ -symmetry operations [15,16]. It is natural that the relationship between drift and peanut deformations viewed from a circular shape is analogous to the 1:2 mode interactions. Letting $z_1 = Qe^{i\phi}$ and $z_2 = Se^{i\psi}$, we rewrite Eq. (9) as

$$\dot{Q} = (M_1 Q^2 + M_2 S^2 + M_3)Q + \beta QS \cos \theta,$$

$$\dot{S} = (N_1 S^2 + N_2 Q^2 + N_3)S + \alpha Q^2 \cos \theta,$$

$$\dot{\theta} = - \left(2\beta S + \frac{\alpha Q^2}{S} \right) \sin \theta, \quad (12)$$

where we set $\theta = \psi - 2\phi$. In addition to the trivial standing disk (SD) spot of $Q=S=0$, we have the fixed points of Eq. (12) with $|\cos \theta|=1$ as the standing peanut (SP) spot of $Q=0$ and $S^2 = -N_3/N_1$. Hereafter we use (M_3, N_3) as the new bifurcation parameter set.

The traveling spot solution of Eq. (13) bifurcates from the SD spot at $M_3=0$ and from the SP spot at $M_3 - M_2 N_3 / N_1 \pm \beta(-N_3 / N_1)^{1/2} = 0$,

$$\begin{aligned} M_1 Q^2 + M_2 S^2 + M_3 \pm \beta S &= 0, \\ (N_1 S^2 + N_2 Q^2 + N_3) S \pm \alpha Q^2 &= 0, \end{aligned} \quad (13)$$

where the traveling spot TS_0 with $\cos \theta = 1$ (TS_π with $\cos \theta = -1$) corresponds to a propagation direction parallel (perpendicular) to the long axis of the deformed shape. The stability and its properties of traveling spot solutions can be obtained by investigating the linearized system of Eq. (9).

As a representative model system fitting our framework, we employ the following activator-substrate-inhibitor reaction-diffusion system:

$$\begin{aligned} u_t &= D_u \Delta u - \frac{uv^2}{1+f_2 w} + f_0(1-u), \\ v_t &= D_v \Delta v + \frac{uv^2}{1+f_2 w} - (f_0 + f_1)v, \\ \tau w_t &= D_w \Delta w + f_3(v-w), \end{aligned} \quad (14)$$

where we set the parameter values $f_0=0.05$, $f_2=0.5$, $f_3=0.2$, $(D_u, D_v, D_w)=(2.0 \times 10^{-4}, 1.0 \times 10^{-4}, 5.0 \times 10^{-4})$, and $\tau=40$ [4]. As shown in Fig. 1(a), by numerical analysis of Eq. (14), we found that the drift and peanut bifurcations occur on the SD branch and the profiles of the associated eigenfunctions are shown in Fig. 1(c). The SP branch appears subcritically at $f_1 \approx 0.0592$ and traveling spot solutions of TS_π and TS_0 emanate from the drift bifurcation points at $f_1 \approx 0.0605$ and 0.0617 on the SP branch, respectively. Especially, the solution profile of TS_π is deformed from peanut shape to disk shape via saddle-node bifurcation. Its stability property changes from saddle to unstable spiral just after the saddle-node point at $f_1 \approx 0.06027$. As shown in Figs. 1(a) and 1(b), TS_π recovers its stability via Hopf bifurcation at $f_1 \approx 0.0634$ and end up with the drift bifurcation point at $f_1 \approx 0.0624$ on the SD branch. We also detect pitchfork bifurcations at $f_1 \approx 0.0632$ and 0.06107 on the TS_π and TS_0 branches; the profiles for the associated eigenfunctions of Ξ_π and Ξ_0 , respectively, show asymmetry perpendicular to the propagation direction as shown in Fig. 1(d). A plausible scenario is that rotational spot solutions for Eq. (14) described later originate in those pitchfork bifurcations. On the other hand, it is confirmed in the reduced ODEs that RS solutions emerge from such bifurcation points as is discussed below. The solution of Eq. (13) becomes unstable when the coefficient of the angle equation of Eq. (12) is positive. That is, the following solutions of Eq. (15) with $|\cos \theta| \neq 1$ emanate via pitchfork bifurcation,

$$Q^2 = \left(-\frac{2\beta}{\alpha} \right) S^2 = \left(-\frac{2\beta}{\alpha} \right) \frac{N_3 + 2M_3}{K},$$

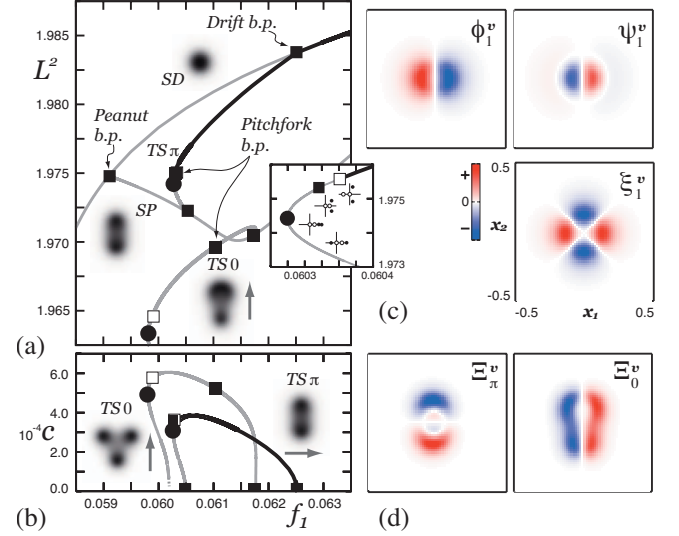


FIG. 1. (Color) (a) Bifurcation diagram of traveling spot solutions in the PDE system of Eq. (11). The solid and gray lines indicate the stable and unstable solutions. The corresponding traveling velocity c is shown in (b). The black and white squares indicate the pitchfork and Hopf points and the black disks show the saddle-node points, respectively. The inset shows the magnified saddle-node point for TS_π branch with eigenvalues distribution. Just before this point, the Hopf and pitchfork bifurcations occur. Note that the tip splitting occurs for TS_0 solutions via a saddle-node bifurcation, and it is expected that its branch ends up to the stationary D_3 spot. (c) Profiles of eigenfunctions of standing disk spot solutions at the bifurcation points: translation-free mode ϕ_1 and deformation vector ψ_1 at the drift bifurcation point; peanut mode ξ_1 at the peanut bifurcation point. (d) Profiles of the symmetry-breaking eigenfunctions of Ξ_π and Ξ_0 at the pitchfork bifurcation points for TS_π and TS_0 solutions. Only the v component is shown here. Spectral computations are done with the system size 1.5×1.5 .

$$\cos^2 \theta = \frac{[N_3(M_2 - 2\beta M_1/\alpha) - M_3(N_1 - 2\beta N_2/\alpha)]^2}{\beta^2(N_3 + 2M_3)K}, \quad (15)$$

where $K = 4\beta M_1/\alpha - 2M_2 - N_1 + 2\beta N_2/\alpha$. Accordingly, we solve the slave part in Eq. (9) as $z_0 = (2/\alpha\beta)^{1/2}(\beta' S e^{i\theta_0} - 1)e^{i\beta S \sin \theta t}/\sin \theta$, where θ_0 is constant. This allows the occurrence of RS motion with radius $|z_0|^2 = 2[(\beta' S)^2 - 1]/(\alpha\beta \sin^2 \theta)$ for $\cos \theta_0 = (\beta' S)^{-1}$. Since the phase speed $\dot{\psi} = 2\dot{\phi} = 2\beta S \sin \theta$ becomes zero at the pitchfork bifurcation point of $|\cos \theta| = 1$, where Q and θ are continuous, clockwise and counterclockwise rotational motions with an infinite radius are equally possible to emanate from a straight motion. Here we consider $\alpha\beta < 0$ which is numerically confirmed as $\alpha \approx -31.8$, $\beta \approx 1.0$, and $\beta' \approx -326.7$ from Eq. (8). The constants of Eqs. (10) and (11) are also computed as $M_1 \approx -61.3$, $M_2 \approx -3.9$, $N_1 \approx -240.0$, and $N_2 \approx -35.6$, for which the eigenfunctions are normalized to satisfy the conditions of Eq. (2). Figures 2(a)–2(c) show the RS solution for the ODE dynamics of Eq. (9), in which the other parameter values are set to $(M_3, N_3) = (0.02, 0.1)$. A bifurcation leading to the onset of RS motion is also shown in

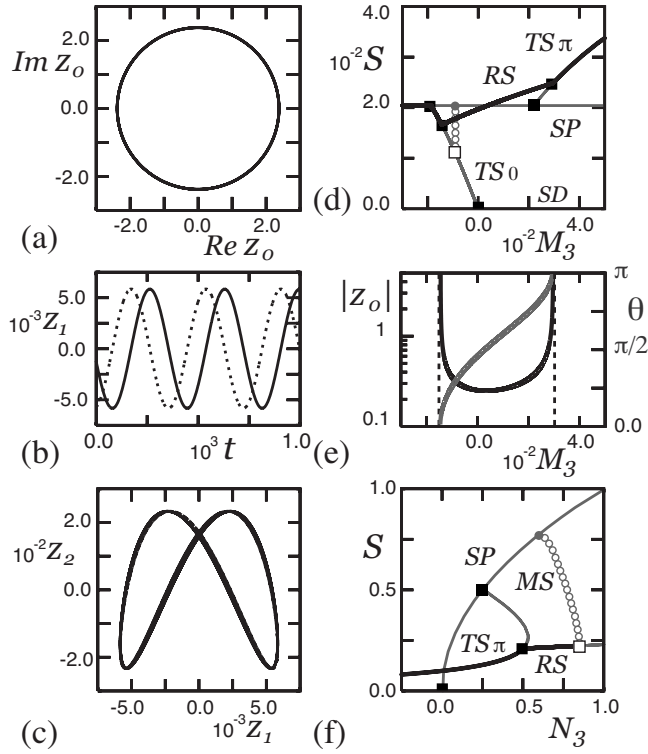


FIG. 2. (a,b,c) 1:2 mode interaction in a rotational spot (RS) motion for ODE of Eq. (6). Real (solid line) and imaginary (dotted line) parts are shown in (b) and (c). (d) Bifurcation diagram of spot solutions for the ODEs of Eq. (9), where N_3 is fixed to 0.1. Stable RS motion appears via pitchfork bifurcations and connects between the TS_0 and TS_π branches. White disk indicates the unstable breather solution. (e) Radius $|z_0|$ (solid line) and angle θ (gray line) change of RS motion. (f) RS motion loses its stability via Torus bifurcation and MS motion (white disk) emanates, where M_3 is fixed to 1.

Figs. 2(d) and 2(e). A stable RS branch emanates from pitchfork bifurcations on the TS branches and connects smoothly between the two types of straight motions of TS_0 and TS_π . The system of Eq. (12) inherits the variety of spot dynamics associated with global behaviors of bifurcation branches, including the hidden unstable branches. We assume that $M_1, M_2, N_1,$ and N_2 are negative and either M_3 or N_3 is positive. Their values can be tuned up to realize the saddle-node structure of the TS_π branch, which is the case for the PDE structure of Fig. 1(a), by analyzing Eq. (13) for $(M_1, M_2, N_1, N_2) = (-8, -2, -1, -10)$ as shown in Fig. 2(f). More systematic calculations of bifurcational structures for 1:2 mode interaction dynamics were carried out by Holmes *et al.* [16,17].

In searching the PDE dynamics for the parameter region close to the saddle-node point in Fig. 1(a), we find the RS motion for $f_1 \in [0.06031; 0.06036]$. Its trajectory of centroid of v -component distribution draws a circle as shown in Fig. 3(a). Figures 3(b) and 3(c) show the spatiotemporal patterns of the v -component profiles along the trace of centroids and circular mapping plot $v(t, \phi)$. A spot almost maintains the shape and rotates with constant velocity. However, there is a small internal breathing motion as the time variation in L^2 slightly oscillates four times during a rotation as shown in

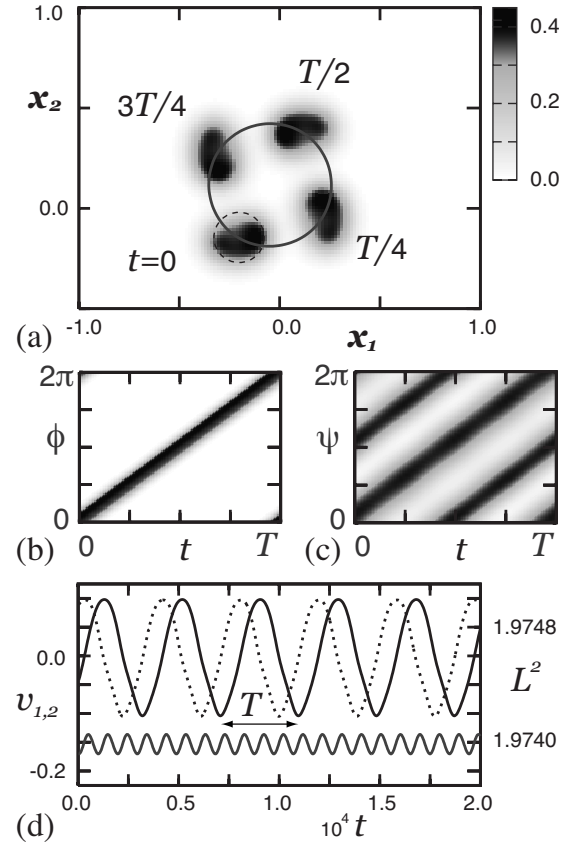


FIG. 3. Rotational spot (RS) motion in the PDE system of Eq. (11). (a) A spot moves in a counterclockwise direction as observed in four superimposed snapshots at $f_1 \approx 0.06035$. The trajectory of the centroid of the v -component distribution is depicted by the solid line. The radius of RS motion is estimated as 0.31. (b) Spatiotemporal pattern of the v -component profile $v(t, \phi)$ along the trace of the centroid. (c) Circular mapping of $v(t, \psi)$ as the radial profile of v component from the centroid, as indicated by the dotted circle in (a). $\psi=0$ is fixed in the x axis and the radius is set to 0.125. (d) Time evolutions of propagation velocity components (v_1, v_2) and L^2 norms are shown by solid, dotted, and gray lines, respectively. We estimate the time periods for RS motion as $T \approx 3.9 \times 10^3$. Computations are carried out with system size 4×4 subject to the periodic boundary condition. The grid sizes are $\Delta x = \Delta y = 2^{-6}$ and $\Delta t = 0.10$.

Fig. 3(d). When f_1 decreases, both radius and period of RS motion decrease. As f_1 continues to decrease, modulatory instability occurs and grows, resulting in a spot splitting behavior. Investigation of the linearization of Eq. (12) shows that the RS solution of Eq. (15) loses its stability via Torus bifurcation and unstable modulatory spot motion, for which both amplitude and phase change in time, may emanate and end up with a heteroclinic bifurcation on the SP branch. The details are left for future work.

In summary, we have studied the localized spot dynamics near the drift and peanut codimension 2 singularity in a class of three-component reaction-diffusion systems. Interaction between the \mathcal{D}_1 and \mathcal{D}_2 symmetry-breaking deformations viewed from a \mathcal{D}_∞ shape of a SD spot is analogous to 1:2 resonance patterns. It turns out that the corresponding bifurcations and the resulting straight motions of TS_0 and TS_π are

crucial for understanding the onset of rotational motion of traveling spots in two dimensions.

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APPENDIX A: CONSTANTS M_1 , M_2 , AND M_3

In the appendix, we will give the derivations of Eqs. (10) and (11). The basic idea is shown in [13], however, it is technically complicated to derive their explicit form for codimension 2 singularity in two dimensions.

Since $(\tau(\mathbf{p})\mathbf{u})_t = -\tau(\mathbf{p})\langle \dot{\mathbf{p}}, \nabla \mathbf{u} \rangle_{L^2}$ holds, we have

$$\begin{aligned} \mathbf{u}_t &= -\tau(\mathbf{p})\langle \dot{\mathbf{p}}, \nabla \mathbf{u} \rangle_{L^2} + \tau(\mathbf{p}) \left\{ \sum_{i=1}^2 \dot{q}_i \boldsymbol{\psi}_i + \sum_{i=1}^2 \dot{s}_i \boldsymbol{\xi}_i \right\} \\ &= \tau(\mathbf{p}) \left\{ -\langle \dot{\mathbf{p}}, \nabla \mathbf{u} \rangle_{L^2} + \sum_{i=1}^2 \dot{q}_i \boldsymbol{\psi}_i + \sum_{i=1}^2 \dot{s}_i \boldsymbol{\xi}_i \right\}, \end{aligned} \quad (\text{A1})$$

where $\langle \dot{\mathbf{p}}, \nabla \mathbf{u} \rangle_{L^2} = \langle \dot{\mathbf{p}}, \nabla S \rangle_{L^2} + q_1 \langle \dot{\mathbf{p}}, \nabla \boldsymbol{\psi}_1 \rangle_{L^2} + q_2 \langle \dot{\mathbf{p}}, \nabla \boldsymbol{\psi}_2 \rangle_{L^2} + s_1 \langle \dot{\mathbf{p}}, \nabla \boldsymbol{\xi}_1 \rangle_{L^2} + s_2 \langle \dot{\mathbf{p}}, \nabla \boldsymbol{\xi}_2 \rangle_{L^2} + \langle \dot{\mathbf{p}}, \nabla \boldsymbol{\zeta}^\dagger \rangle_{L^2}$ and

$$\begin{aligned} \mathcal{L}(\mathbf{u}) + \sum_{i=1}^2 \eta_i \mathbf{g}_i(\mathbf{u}) \\ = \tau(P) \left\{ -\sum_{i=1}^2 q_i \phi_i + L \boldsymbol{\zeta}^\dagger + \frac{1}{2} F''(S) \mathbf{W}^2 + \frac{1}{6} F'''(S) \mathbf{W}^3 \right. \\ \left. + \sum_{i=1}^2 \eta_i \mathbf{g}_i(S) + \sum_{i=1}^2 \eta_i \mathbf{g}'_i(S) \mathbf{W} \right\}, \end{aligned} \quad (\text{A2})$$

where $\mathbf{W} = \sum_{i=1}^2 q_i \boldsymbol{\psi}_i(\mathbf{r}) + \sum_{i=1}^2 s_i \boldsymbol{\xi}_i(\mathbf{r}) + \boldsymbol{\zeta}^\dagger$. Here we will convert the equation of Eq. (1) of \mathbf{u} to that of $(\mathbf{p}, \mathbf{q}, \mathbf{s})$. Taking the inner products with $\boldsymbol{\psi}_i^*$ ($i=1, 2$), we have

$$\begin{aligned} \langle \mathbf{u}_t, \boldsymbol{\psi}_1^* \rangle_{L^2} &= -\dot{p}_1 \langle \boldsymbol{\phi}_1, \boldsymbol{\psi}_1^* \rangle_{L^2} - \dot{p}_1 s_1 \langle \boldsymbol{\xi}_{1x_1}, \boldsymbol{\psi}_1^* \rangle_{L^2} \\ &\quad - \dot{p}_2 s_2 \langle \boldsymbol{\xi}_{2x_2}, \boldsymbol{\psi}_1^* \rangle_{L^2}, \end{aligned}$$

$$\begin{aligned} \left\langle \mathcal{L}(\mathbf{u}) + \sum_{i=1}^2 \eta_i \mathbf{g}_i(\mathbf{u}), \boldsymbol{\psi}_1^* \right\rangle_{L^2} &= -q_1 \langle \boldsymbol{\phi}_1, \boldsymbol{\psi}_1^* \rangle_{L^2} \\ &\quad + q_1 s_1 \langle L \boldsymbol{\zeta}_7 + F''(S) \boldsymbol{\psi}_1 \boldsymbol{\xi}_1, \boldsymbol{\psi}_1^* \rangle_{L^2} \\ &\quad + q_2 s_2 \langle L \boldsymbol{\zeta}_8 + F''(S) \boldsymbol{\psi}_2 \boldsymbol{\xi}_2, \boldsymbol{\psi}_1^* \rangle_{L^2}, \end{aligned}$$

$$\begin{aligned} \langle \mathbf{u}_t, \boldsymbol{\psi}_2^* \rangle_{L^2} &= -\dot{p}_2 \langle \boldsymbol{\phi}_2, \boldsymbol{\psi}_2^* \rangle_{L^2} - \dot{p}_1 s_2 \langle \boldsymbol{\xi}_{2x_1}, \boldsymbol{\psi}_2^* \rangle_{L^2} \\ &\quad - \dot{p}_2 s_1 \langle \boldsymbol{\xi}_{1x_2}, \boldsymbol{\psi}_2^* \rangle_{L^2}, \end{aligned}$$

$$\begin{aligned} \left\langle \mathcal{L}(\mathbf{u}) + \sum_{i=1}^2 \eta_i \mathbf{g}_i(\mathbf{u}), \boldsymbol{\psi}_2^* \right\rangle_{L^2} &= -q_2 \langle \boldsymbol{\phi}_2, \boldsymbol{\psi}_2^* \rangle_{L^2} \\ &\quad + q_1 s_2 \langle L \boldsymbol{\zeta}_9 + F''(S) \boldsymbol{\psi}_1 \boldsymbol{\xi}_2, \boldsymbol{\psi}_2^* \rangle_{L^2} \\ &\quad + q_2 s_1 \langle L \boldsymbol{\zeta}_{10} \\ &\quad + F''(S) \boldsymbol{\psi}_2 \boldsymbol{\xi}_1, \boldsymbol{\psi}_2^* \rangle_{L^2}, \end{aligned} \quad (\text{A3})$$

where we show only nonzero terms. Since $\boldsymbol{\psi}_1 = \cos \theta \psi(r)$, $\boldsymbol{\phi}_1 = \cos \theta \phi(r)$, and $\boldsymbol{\xi}_1 = \cos 2\theta \xi(r)$ and so on, we can rewrite Eq. (8) as

$$\begin{aligned} \left\langle F''(S) \psi^2 + 2 \left(\psi_r - \frac{\psi}{r} \right) - 2\alpha \xi, \xi^* \right\rangle &= 0, \\ \left\langle F''(S) \psi \xi + \xi_r + \frac{2\xi}{r} - \beta \psi - \beta' \phi, \phi^* \right\rangle &= 0, \\ \left\langle F''(S) \psi \xi + \xi_r + \frac{2\xi}{r} - \beta \psi - \beta' \phi, \psi^* \right\rangle &= 0. \end{aligned} \quad (\text{A4})$$

We know that $\dot{p}_1 = q_1 - \beta'(q_1 s_1 + q_2 s_2)$ and $\dot{p}_2 = q_2 - \beta'(q_1 s_2 - q_2 s_1)$, respectively. Next, in order to obtain the equation of motion of q_i , we take the inner products with $\boldsymbol{\phi}_i^*$ ($i=1, 2$) as

$$\begin{aligned} \langle \mathbf{u}_t, \boldsymbol{\phi}_1^* \rangle_{L^2} &= -\dot{p}_1 \langle \boldsymbol{\zeta}_{x_1}^\dagger, \boldsymbol{\phi}_1^* \rangle_{L^2} + \dot{q}_1 \langle \boldsymbol{\psi}_1, \boldsymbol{\phi}_1^* \rangle_{L^2} \\ &= \left\langle \mathcal{L}(\mathbf{u}) + \sum_{i=1}^2 \eta_i \mathbf{g}_i(\mathbf{u}), \boldsymbol{\phi}_1^* \right\rangle_{L^2}, \\ \langle \mathbf{u}_t, \boldsymbol{\phi}_2^* \rangle_{L^2} &= -\dot{p}_2 \langle \boldsymbol{\zeta}_{x_2}^\dagger, \boldsymbol{\phi}_2^* \rangle_{L^2} + \dot{q}_2 \langle \boldsymbol{\psi}_2, \boldsymbol{\phi}_2^* \rangle_{L^2} \\ &= \left\langle \mathcal{L}(\mathbf{u}) + \sum_{i=1}^2 \eta_i \mathbf{g}_i(\mathbf{u}), \boldsymbol{\phi}_2^* \right\rangle_{L^2}. \end{aligned} \quad (\text{A5})$$

Computing each term of Eq. (A3), the first term of M_1 is given as

$$\begin{aligned} \frac{1}{6} \langle F'''(S) \boldsymbol{\psi}_1^3, \boldsymbol{\phi}_1^* \rangle_{L^2} &= \frac{1}{6} \int_0^{2\pi} \cos^4 \theta d\theta \int_0^\infty r F'''(S) \psi^3 \phi^* dr \\ &= \frac{\pi}{8} \int_0^\infty r F'''(S) \psi^3 \phi^* dr. \end{aligned} \quad (\text{A6})$$

By similar calculations to the above, we have

$$\begin{aligned} \frac{1}{6} \langle F'''(S) \boldsymbol{\psi}_1^3, \boldsymbol{\phi}_1^* \rangle_{L^2} &= \frac{1}{2} \langle F'''(S) \boldsymbol{\psi}_1 \boldsymbol{\psi}_2^2, \boldsymbol{\phi}_1^* \rangle_{L^2} \\ &= \frac{1}{6} \langle F'''(S) \boldsymbol{\psi}_2^3, \boldsymbol{\phi}_2^* \rangle_{L^2} \\ &= \frac{1}{2} \langle F'''(S) \boldsymbol{\psi}_1^2 \boldsymbol{\psi}_2, \boldsymbol{\phi}_2^* \rangle_{L^2} \equiv \pi M'_1. \end{aligned} \quad (\text{A7})$$

We can rewrite Eq. (4) as

$$L \boldsymbol{\zeta}_1 + \frac{1}{2} F''(S) \cos^2 \theta \psi^2 + \cos^2 \theta \psi_r + \sin^2 \theta \frac{\psi}{r} = \alpha \cos 2\theta \xi,$$

$$\begin{aligned}
 L\zeta_2 + \frac{1}{2}F''(S)\sin^2\theta\psi^2 + \sin^2\theta\psi_r + \cos^2\theta\frac{\psi}{r} &= -\alpha\cos 2\theta\xi, \\
 L\zeta_3 + F''(S)\sin\theta\cos\theta\psi^2 + 2\sin\theta\cos\theta\left(\psi_r - \frac{\psi}{r}\right) & \\
 &= 2\alpha\sin 2\theta\xi. \tag{A8}
 \end{aligned}$$

The term of $\zeta_1 - \zeta_2$ satisfies

$$\begin{aligned}
 -L(\zeta_1 - \zeta_2) &= \frac{1}{2}(\cos^2\theta - \sin^2\theta)\left[F''(S)\psi^2 + 2\left(\psi_r - \frac{\psi}{r}\right)\right] \\
 &\quad - 2\alpha\cos 2\theta\xi \\
 &= \frac{1}{2}\sin\left(2\theta + \frac{\pi}{2}\right)\left[F''(S)\psi^2 + 2\left(\psi_r - \frac{\psi}{r}\right) - 4\alpha\xi\right]. \tag{A9}
 \end{aligned}$$

Hence we have $(\zeta_1 - \zeta_2)(r, \theta) = \zeta_3(r, \theta + \frac{\pi}{4})$. We note that $\zeta_1 = \cos^2\theta\tilde{\zeta}_1(r) + \sin^2\theta\tilde{\zeta}_2(r)$, $\zeta_2 = \sin^2\theta\tilde{\zeta}_1(r) + \cos^2\theta\tilde{\zeta}_2(r)$, and $\zeta_3 = \sin 2\theta\tilde{\zeta}_3(r)$ hold. It is easy to see that $\zeta_2(r, \theta) = \zeta_1(r, \theta + \frac{\pi}{2})$. Thus, the second term of M_1 has the following:

$$\begin{aligned}
 \langle F''(S)\psi_1\zeta_1, \phi_1^* \rangle_{L^2} &= \int_0^{2\pi} \int_0^\infty \cos^2\theta r \langle F''(S)\psi\zeta_1, \phi^* \rangle dr d\theta \\
 &= \frac{3\pi}{4} \int_0^\infty r \langle F''\psi\tilde{\zeta}_1, \phi^* \rangle dr \\
 &\quad + \frac{\pi}{4} \int_0^\infty r \langle F''\psi\tilde{\zeta}_2, \phi^* \rangle dr. \tag{A10}
 \end{aligned}$$

Similarly, we have

$$\begin{aligned}
 \langle F''(S)\psi_1\zeta_1, \phi_1^* \rangle_{L^2} &= \langle F''(S)\psi_1\zeta_2, \phi_1^* \rangle_{L^2} + \langle F''(S)\psi_2\zeta_3, \phi_1^* \rangle_{L^2} \\
 &= \langle F''(S)\psi_2\zeta_2, \phi_2^* \rangle_{L^2} \\
 &= \langle F''(S)\psi_2\zeta_1, \phi_2^* \rangle_{L^2} + \langle F''(S)\psi_1\zeta_3, \phi_2^* \rangle_{L^2} \\
 &\equiv \pi M_1', \tag{A11}
 \end{aligned}$$

where we use the relation of $\tilde{\zeta}_1(r) - \tilde{\zeta}_2(r) = \tilde{\zeta}_3(r)$. The last term of M_1 is obtained as

$$\begin{aligned}
 \langle \zeta_{1x_1}, \phi_1^* \rangle_{L^2} &= \int_0^{2\pi} \int_0^\infty \cos^2\theta r \langle \zeta_{1r}, \phi^* \rangle dr d\theta \\
 &\quad - \int_0^{2\pi} \int_0^\infty \sin\theta\cos\theta \langle \zeta_{1\theta}, \phi^* \rangle dr d\theta \\
 &= \frac{3\pi}{4} \int_0^\infty r \langle \tilde{\zeta}_{1r}, \phi^* \rangle dr + \frac{\pi}{4} \int_0^\infty r \langle \tilde{\zeta}_{2r}, \phi^* \rangle dr \\
 &\quad - \frac{\pi}{2} \int_0^\infty \langle \tilde{\zeta}_2 - \tilde{\zeta}_1, \phi^* \rangle dr. \tag{A12}
 \end{aligned}$$

Here, we also have

$$\begin{aligned}
 &\langle \zeta_{1x_1}, \phi_1^* \rangle_{L^2} - \langle \zeta_{2x_1}, \phi_1^* \rangle_{L^2} - \langle \zeta_{3x_2}, \phi_1^* \rangle_{L^2} \\
 &= \frac{\pi}{2} \int_0^\infty r \langle \tilde{\zeta}_{1r}, \phi^* \rangle dr - \frac{\pi}{2} \int_0^\infty r \langle \tilde{\zeta}_{2r}, \phi^* \rangle dr \\
 &\quad + \pi \int_0^\infty \langle \tilde{\zeta}_1 - \tilde{\zeta}_2, \phi^* \rangle dr - \frac{\pi}{2} \int_0^\infty r \langle \tilde{\zeta}_{3r}, \phi^* \rangle dr \\
 &\quad - \pi \int_0^\infty \langle \tilde{\zeta}_3, \phi^* \rangle dr = 0. \tag{A13}
 \end{aligned}$$

In view of the above results, we have

$$\begin{aligned}
 \langle \zeta_{1x_1}, \phi_1^* \rangle_{L^2} &= \langle \zeta_{2x_1}, \phi_1^* \rangle_{L^2} + \langle \zeta_{3x_2}, \phi_1^* \rangle_{L^2} \\
 &= \langle \zeta_{2x_2}, \phi_2^* \rangle_{L^2} \\
 &= \langle \zeta_{1x_2}, \phi_2^* \rangle_{L^2} + \langle \zeta_{3x_1}, \phi_2^* \rangle_{L^2} \\
 &\equiv \pi M_1'''. \tag{A14}
 \end{aligned}$$

In this way, we obtain $M_1 = M_1' + M_1'' + M_1'''$.

Let us now rewrite Eq. (5) as

$$L\zeta_4 + \frac{1}{2}F''(S)\cos^2 2\theta\xi^2 = 0,$$

$$L\zeta_5 + \frac{1}{2}F''(S)\sin^2 2\theta\xi^2 = 0,$$

$$L\zeta_6 + F''(S)\sin 2\theta\cos 2\theta\xi^2 = 0. \tag{A15}$$

We note that $\zeta_4 = \cos^2 2\theta\tilde{\zeta}_4$, $\zeta_5 = \sin^2 2\theta\tilde{\zeta}_5$, and $\zeta_6 = \sin 4\theta\tilde{\zeta}_6$. It is easy to see that $\zeta_5(r, \theta) = \zeta_4(r, \theta + \frac{\pi}{4})$. The term of $\zeta_4 - \zeta_5$ satisfies

$$\begin{aligned}
 -L(\zeta_4 - \zeta_5) &= \frac{1}{2}(\cos^2 2\theta - \sin^2 2\theta)F''(S)\xi^2 \\
 &= \frac{1}{2}\sin\left(4\theta + \frac{\pi}{2}\right)F''(S)\xi^2.
 \end{aligned}$$

Hence we have $(\zeta_4 - \zeta_5)(r, \theta) = \zeta_6(r, \theta + \frac{\pi}{8})$. We can rewrite Eq. (6) as

$$\begin{aligned}
 L\zeta_7 + F''(S)\cos\theta\cos 2\theta\psi\xi + \cos\theta\cos 2\theta\xi_r + \sin\theta\sin 2\theta\frac{2\xi}{r} & \\
 &= \cos\theta(\beta\psi + \beta'\phi),
 \end{aligned}$$

$$\begin{aligned}
 L\zeta_8 + F''(S)\sin\theta\sin 2\theta\psi\xi + \sin\theta\sin 2\theta\xi_r + \cos\theta\cos 2\theta\frac{2\xi}{r} & \\
 &= \cos\theta(\beta\psi + \beta'\phi),
 \end{aligned}$$

$$\begin{aligned}
 L\zeta_9 + F''(S)\cos\theta\sin 2\theta\psi\xi + \cos\theta\sin 2\theta\xi_r - \sin\theta\cos 2\theta\frac{2\xi}{r} & \\
 &= \sin\theta(\beta\psi + \beta'\phi),
 \end{aligned}$$

$$L\zeta_{10} + F''(S)\sin\theta\cos2\theta\psi\xi + \sin\theta\cos2\theta\xi_r - \cos\theta\sin2\theta\frac{2\xi}{r} = -\sin\theta(\beta\psi + \beta'\phi). \quad (\text{A16})$$

We note that $\zeta_7 = \cos\theta\cos2\theta\tilde{\zeta}_7 + \sin\theta\sin2\theta\tilde{\zeta}_8$, $\zeta_8 = \sin\theta\sin2\theta\tilde{\zeta}_7 + \cos\theta\cos2\theta\tilde{\zeta}_8$, $\zeta_9 = \cos\theta\sin2\theta\tilde{\zeta}_9 + \sin\theta\cos2\theta\tilde{\zeta}_{10}$, and $\zeta_{10} = \sin\theta\cos2\theta\tilde{\zeta}_9 + \cos\theta\sin2\theta\tilde{\zeta}_{10}$. The terms of $\zeta_7 \pm \zeta_8$ satisfy

$$\begin{aligned} -L(\zeta_7 + \zeta_8) &= (\cos\theta\cos2\theta + \sin\theta\sin2\theta) \\ &\times \left(F''(S)\psi\xi + \xi_r - \frac{2\xi}{r} \right) - 2\cos\theta(\beta\psi + \beta'\phi) \\ &= \cos\theta \left(F''(S)\psi\xi + \xi_r - \frac{2\xi}{r} - 2(\beta\psi + \beta'\phi) \right), \end{aligned} \quad (\text{A17})$$

$$\begin{aligned} -L(\zeta_7 - \zeta_8) &= (\cos\theta\cos2\theta - \sin\theta\sin2\theta) \\ &\times \left(F''(S)\psi\xi + \xi_r - \frac{2\xi}{r} \right) \\ &= \cos3\theta \left(F''(S)\psi\xi + \xi_r - \frac{2\xi}{r} \right), \end{aligned} \quad (\text{A18})$$

and the terms of $\zeta_9 \pm \zeta_{10}$ satisfy

$$\begin{aligned} -L(\zeta_9 + \zeta_{10}) &= (\cos\theta\sin2\theta + \sin\theta\cos2\theta) \\ &\times \left(F''(S)\psi\xi + \xi_r - \frac{2\xi}{r} \right) \\ &= \sin3\theta \left(F''(S)\psi\xi + \xi_r - \frac{2\xi}{r} \right), \end{aligned} \quad (\text{A19})$$

$$\begin{aligned} -L(\zeta_9 - \zeta_{10}) &= (\cos\theta\sin2\theta - \sin\theta\cos2\theta) \\ &\times \left(F''(S)\psi\xi + \xi_r - \frac{2\xi}{r} \right) - 2\sin\theta(\beta\psi + \beta'\phi) \\ &= \sin\theta \left(F''(S)\psi\xi + \xi_r - \frac{2\xi}{r} - 2(\beta\psi + \beta'\phi) \right). \end{aligned} \quad (\text{A20})$$

Hence we have $(\zeta_7 + \zeta_8)(r, \theta) = (\zeta_9 - \zeta_{10})(r, \theta + \frac{\pi}{2})$ and $(\zeta_7 - \zeta_8)(r, \theta) = (\zeta_9 + \zeta_{10})(r, \theta + \frac{\pi}{6})$. The first three terms of M_2 are given as

$$\begin{aligned} &\left\langle \frac{1}{2}F'''(S)\xi_1^2\psi_1 + F''(S)\psi_1\zeta_4 + F''(S)\xi_1\zeta_7, \Phi_1^* \right\rangle_{L^2} \\ &= \frac{1}{2}\int_0^{2\pi}\cos^2\theta\cos^22\theta d\theta\int_0^\infty r\langle F'''(S)\xi^2\psi, \Phi^* \rangle dr \\ &+ \int_0^{2\pi}\cos^2\theta\cos^22\theta d\theta\int_0^\infty r\langle F''(S)\tilde{\zeta}_4\psi, \Phi^* \rangle dr \\ &+ \int_0^{2\pi}\cos^2\theta\cos^22\theta d\theta\int_0^\infty r\langle F''(S)\tilde{\zeta}_7\xi, \Phi^* \rangle dr \end{aligned}$$

$$\begin{aligned} &+ \int_0^{2\pi}\sin\theta\cos\theta\sin2\theta\cos2\theta d\theta\int_0^\infty r\langle F''(S)\tilde{\zeta}_8\xi, \Phi^* \rangle dr \\ &= \frac{\pi}{4}\int_0^\infty r\langle F'''(S)\xi^2\psi, \Phi^* \rangle dr + \frac{\pi}{2}\int_0^\infty r\langle F''(S)\tilde{\zeta}_4\psi, \Phi^* \rangle dr \\ &+ \frac{\pi}{2}\int_0^\infty r\langle F''(S)\tilde{\zeta}_7\xi, \Phi^* \rangle dr. \end{aligned} \quad (\text{A21})$$

Quite similarly, we have

$$\begin{aligned} &\left\langle \frac{1}{2}F'''(S)\xi_1^2\psi_1 + F''(S)\psi_1\zeta_4 + F''(S)\xi_1\zeta_7, \Phi_1^* \right\rangle_{L^2} \\ &= \left\langle \frac{1}{2}F'''(S)\xi_1^2\psi_2 + F''(S)\psi_2\zeta_4 + F''(S)\xi_1\zeta_{10}, \Phi_2^* \right\rangle_{L^2} \\ &= \left\langle \frac{1}{2}F'''(S)\xi_2^2\psi_2 + F''(S)\psi_2\zeta_5 + F''(S)\xi_2\zeta_8, \Phi_2^* \right\rangle_{L^2} \\ &= \left\langle \frac{1}{2}F'''(S)\xi_2^2\psi_1 + F''(S)\psi_1\zeta_5 + F''(S)\xi_2\zeta_9, \Phi_1^* \right\rangle_{L^2} \equiv \pi M'_2. \end{aligned} \quad (\text{A22})$$

Here we use $\tilde{\zeta}_4(r) = \tilde{\zeta}_5(r) = \tilde{\zeta}_6(r)$ and $\tilde{\zeta}_7(r) = \tilde{\zeta}_9(r)$ and $\tilde{\zeta}_8(r) = -\tilde{\zeta}_{10}(r)$. The last two terms of M_2 are obtained as

$$\begin{aligned} \langle \zeta_{4x_1}, \Phi_1^* \rangle_{L^2} &= \int_0^{2\pi}\cos^2\theta\cos^22\theta d\theta\int_0^\infty r\langle \tilde{\zeta}_{4r}, \Phi^* \rangle dr \\ &+ 4\int_0^{2\pi}\sin\theta\cos\theta\sin2\theta\cos2\theta d\theta \\ &\times \int_0^\infty \langle \tilde{\zeta}_4, \Phi^* \rangle dr \\ &= \frac{\pi}{2}\int_0^\infty r\langle \tilde{\zeta}_{4r}, \Phi^* \rangle dr \end{aligned} \quad (\text{A23})$$

and

$$\begin{aligned} \langle \xi_{1x_1}, \Phi_1^* \rangle_{L^2} &= \int_0^{2\pi}\cos^2\theta\cos2\theta d\theta\int_0^\infty r\langle \xi_r, \Phi^* \rangle dr \\ &+ 2\int_0^{2\pi}\sin\theta\cos\theta\sin2\theta d\theta\int_0^\infty \langle \xi, \Phi^* \rangle dr \\ &= \frac{\pi}{2}\int_0^\infty r\langle \xi_r, \Phi^* \rangle dr + \pi\int_0^\infty \langle \xi, \Phi^* \rangle dr. \end{aligned} \quad (\text{A24})$$

By similar calculations to the above, we have

$$\langle \zeta_{4x_1}, \Phi_1^* \rangle_{L^2} = \langle \zeta_{5x_2}, \Phi_2^* \rangle_{L^2} = \langle \zeta_{4x_2}, \Phi_2^* \rangle_{L^2} = \langle \zeta_{5x_1}, \Phi_1^* \rangle_{L^2} \equiv \pi M''_2 \quad (\text{A25})$$

and

$$\begin{aligned}
 \langle \xi_{1x_1}, \phi_1^* \rangle_{L^2} &= \langle \xi_{2x_1}, \phi_2^* \rangle_{L^2} \\
 &= \langle \xi_{2x_2}, \phi_1^* \rangle_{L^2} \\
 &= - \langle \xi_{1x_2}, \phi_2^* \rangle_{L^2} \\
 &\equiv \pi M_2'''.
 \end{aligned} \tag{A26}$$

In this way, we obtain $M_2 = M_2' + M_2'' - \beta' M_2'''$. The terms of M_3 are shown quite similarly as

$$\begin{aligned}
 &\langle F''(S) \psi_1 \zeta_{11}, \phi_1^* \rangle_{L^2} + \langle g_1'(S) \psi_1, \phi_1^* \rangle_{L^2} \\
 &= \int_0^{2\pi} \cos^2 \theta d\theta \int_0^\infty r \langle F''(S) \psi \zeta_{11}, \phi^* \rangle dr \\
 &\quad + \int_0^{2\pi} \cos^2 \theta d\theta \int_0^\infty r \langle g_1'(S) \psi, \phi^* \rangle dr \\
 &= \pi \int_0^\infty r \langle F''(S) \tilde{\zeta}_{11}, \phi^* \rangle dr + \pi \int_0^\infty r \langle g_1'(S) \psi, \phi^* \rangle dr \\
 &= \langle F''(S) \psi_2 \zeta_{11}, \phi_2^* \rangle_{L^2} + \langle g_1'(S) \psi_2, \phi_2^* \rangle_{L^2} \equiv \pi M_3' \tag{A27}
 \end{aligned}$$

and

$$\begin{aligned}
 &\langle F''(S) \psi_1 \zeta_{12}, \phi_1^* \rangle_{L^2} + \langle g_2'(S) \psi_1, \phi_1^* \rangle_{L^2} \\
 &= \int_0^{2\pi} \cos^2 \theta d\theta \int_0^\infty r \langle F''(S) \psi \zeta_{12}, \phi^* \rangle dr \\
 &\quad + \int_0^{2\pi} \cos^2 \theta d\theta \int_0^\infty r \langle g_2'(S) \psi, \phi^* \rangle dr \\
 &= \pi \int_0^\infty r \langle F''(S) \tilde{\zeta}_{12}, \phi^* \rangle dr + \pi \int_0^\infty r \langle g_2'(S) \psi, \phi^* \rangle dr \\
 &= \langle F''(S) \psi_2 \zeta_{12}, \phi_2^* \rangle_{L^2} + \langle g_2'(S) \psi_2, \phi_2^* \rangle_{L^2} \equiv \pi M_3''.
 \end{aligned} \tag{A28}$$

Here ζ_{11} and ζ_{12} are radially symmetric as $\zeta_{11} = \tilde{\zeta}_{11}(r)$ and $\zeta_{12} = \tilde{\zeta}_{12}(r)$, respectively. The last term to M_3 is obtained as

$$\begin{aligned}
 \langle \xi_{1x_1}, \phi_1^* \rangle_{L^2} &= \int_0^{2\pi} \cos^2 \theta d\theta \int_0^\infty r \langle \xi_{11r}, \phi^* \rangle dr \\
 &= \pi \int_0^\infty r \langle \tilde{\xi}_{11r}, \phi^* \rangle dr \\
 &= \langle \xi_{11x_2}, \phi_2^* \rangle_{L^2} \\
 &\equiv \pi M_3'''.
 \end{aligned} \tag{A29}$$

and

$$\begin{aligned}
 \langle \xi_{12x_1}, \phi_1^* \rangle_{L^2} &= \int_0^{2\pi} \cos^2 \theta d\theta \int_0^\infty r \langle \xi_{12r}, \phi^* \rangle dr \\
 &= \pi \int_0^\infty r \langle \tilde{\xi}_{12r}, \phi^* \rangle dr \\
 &= \langle \xi_{12x_2}, \phi_2^* \rangle_{L^2} \\
 &\equiv \pi M_3'''.
 \end{aligned} \tag{A30}$$

We arrive at the results of $M_3 = \eta_1(M_3' + M_3'') + \eta_2(M_3''' + M_3''')$. It is remarked that, for numerical computations of the constants, we shall normalize eigenfunctions of $\psi_i^0, \psi_i^{*0}, \phi_i^{*0}$ obtained from numerical spectral analysis. According to Eq. (2), the eigenfunctions are given by

$$\psi_i = \psi_i^0 - \frac{\langle \phi_i, \psi_i^0 \rangle_{L^2}}{\langle \phi_i, \phi_i \rangle_{L^2}} \phi_i,$$

$$\phi_i^* = \frac{\pi}{\langle \phi_i, \psi_i^{*0} \rangle_{L^2}} \phi_i^{*0},$$

$$\begin{aligned}
 \psi_i^* &= \frac{\pi}{\langle \phi_i, \psi_i^{*0} \rangle_{L^2}} \psi_i^{*0} + \frac{\pi}{\langle \psi_i^0, \phi_i^{*0} \rangle_{L^2}} \left(\frac{\langle \phi_i, \psi_i^0 \rangle_{L^2}}{\langle \phi_i, \phi_i \rangle_{L^2}} \right. \\
 &\quad \left. - \frac{\langle \psi_i^0, \psi_i^{*0} \rangle_{L^2}}{\langle \phi_i, \psi_i^{*0} \rangle_{L^2}} \right) \phi_i^{*0}.
 \end{aligned} \tag{A31}$$

APPENDIX B: CONSTANTS $N_1, N_2,$ AND N_3

By similar calculations to those shown in the previous subsection, we have

$$\begin{aligned}
 &\langle F''(S) \xi_1 \zeta_{11}, \xi_1^* \rangle_{L^2} + \langle g_1'(S) \xi_1, \xi_1^* \rangle_{L^2} \\
 &= \int_0^{2\pi} \cos^2 2\theta d\theta \int_0^\infty r \langle F''(S) \xi \zeta_{11}, \xi^* \rangle dr \\
 &\quad + \int_0^{2\pi} \cos^2 2\theta d\theta \int_0^\infty r \langle g_1'(S) \xi, \xi^* \rangle dr \\
 &= \pi \int_0^\infty r \langle F''(S) \tilde{\xi} \zeta_{11}, \xi^* \rangle dr + \pi \int_0^\infty r \langle g_1'(S) \xi, \xi^* \rangle dr \\
 &= \langle F''(S) \xi_2 \zeta_{11}, \xi_2^* \rangle_{L^2} + \langle g_1'(S) \xi_2, \xi_2^* \rangle_{L^2} \\
 &\equiv \pi N_3'
 \end{aligned} \tag{B1}$$

and

$$\begin{aligned}
 & \langle F''(S)\xi_1\zeta_{12}, \xi_1^* \rangle_{L^2} + \langle g_2'(S)\xi_1, \xi_1^* \rangle_{L^2} \\
 &= \int_0^{2\pi} \cos^2 2\theta d\theta \int_0^\infty r \langle F''(S)\xi\xi_{12}, \xi^* \rangle dr \\
 &+ \int_0^{2\pi} \cos^2 2\theta d\theta \int_0^\infty r \langle g_2'(S)\xi, \xi^* \rangle dr \\
 &= \pi \int_0^\infty r \langle F''(S)\xi\tilde{\xi}_{12}, \xi^* \rangle dr + \pi \int_0^\infty r \langle g_2'(S)\xi, \xi^* \rangle dr \\
 &= \langle F''(S)\xi_2\zeta_{12}, \xi_2^* \rangle_{L^2} + \langle g_2'(S)\xi_2, \xi_2^* \rangle_{L^2} \\
 &\equiv \pi N_3'.
 \end{aligned} \tag{B2}$$

Here we take the inner product with ξ_i^* ($i=1,2$) and obtain $N_3 = \eta_1 N_3' + \eta_2 N_3''$. The first term of N_1 is given as

$$\begin{aligned}
 \frac{1}{6} \langle F'''(S)\xi_1^3, \xi_1^* \rangle_{L^2} &= \frac{1}{6} \int_0^{2\pi} \cos^4 2\theta d\theta \int_0^\infty r \langle F'''(S)\xi^3, \xi^* \rangle dr \\
 &= \frac{\pi}{8} \int_0^\infty r \langle F'''(S)\xi^3, \xi^* \rangle dr.
 \end{aligned} \tag{B3}$$

Moreover, we have

$$\begin{aligned}
 \frac{1}{6} \langle F'''(S)\xi_1^3, \xi_1^* \rangle_{L^2} &= \frac{1}{2} \langle F'''(S)\xi_1\xi_2^2, \xi_1^* \rangle_{L^2} = \frac{1}{6} \langle F'''(S)\xi_2^3, \xi_2^* \rangle_{L^2} \\
 &= \frac{1}{2} \langle F'''(S)\xi_1^2\xi_2, \xi_2^* \rangle_{L^2} \equiv \pi N_1'.
 \end{aligned} \tag{B4}$$

We show the second term of N_1 as

$$\begin{aligned}
 \langle F''(S)\xi_1\zeta_4, \xi_1^* \rangle_{L^2} &= \int_0^{2\pi} \cos^4 2\theta d\theta \int_0^\infty r \langle F''(S)\xi\tilde{\xi}_4, \xi^* \rangle dr \\
 &= \frac{3\pi}{4} \int_0^\infty r \langle F''(S)\xi\tilde{\xi}_4, \xi^* \rangle dr.
 \end{aligned} \tag{B5}$$

By similar calculations to the above, we have

$$\begin{aligned}
 \langle F''(S)\xi_1\zeta_4, \xi_1^* \rangle_{L^2} &= \langle F''(S)\xi_1\zeta_5, \xi_1^* \rangle_{L^2} + \langle F''(S)\xi_2\zeta_6, \xi_1^* \rangle_{L^2} \\
 &= \langle F''(S)\xi_2\zeta_5, \xi_2^* \rangle_{L^2} = \langle F''(S)\xi_1\zeta_6, \xi_2^* \rangle_{L^2} \\
 &+ \langle F''(S)\xi_2\zeta_4, \xi_2^* \rangle_{L^2} \equiv \pi N_1''.
 \end{aligned} \tag{B6}$$

In this way, we obtain $N_1 = N_1' + N_1''$.

Last, we shall consider the first three terms of N_2 as

$$\begin{aligned}
 & \left\langle \frac{1}{2} F'''(S)\psi_1^2\xi_1 + F''(S)\psi_1\zeta_7 + F''(S)\xi_1\zeta_1, \xi_1^* \right\rangle_{L^2} \\
 &= \frac{1}{2} \int_0^{2\pi} \cos^2 \theta \cos^2 2\theta d\theta \int_0^\infty r \langle F'''(S)\psi^2\xi, \xi^* \rangle dr + \int_0^{2\pi} \cos^2 \theta \cos^2 2\theta d\theta \int_0^\infty r \langle F''(S)\tilde{\zeta}_7\psi, \xi^* \rangle dr \\
 &+ \int_0^{2\pi} \sin \theta \cos \theta \sin 2\theta \cos 2\theta d\theta \int_0^\infty r \langle F''(S)\tilde{\zeta}_8\psi, \xi^* \rangle dr + \int_0^{2\pi} \cos^2 \theta \cos^2 2\theta d\theta \int_0^\infty r \langle F''(S)\tilde{\zeta}_1\xi, \xi^* \rangle dr \\
 &+ \int_0^{2\pi} \sin^2 \theta \cos^2 2\theta d\theta \int_0^\infty r \langle F''(S)\tilde{\zeta}_2\xi, \xi^* \rangle dr \\
 &= \frac{\pi}{4} \int_0^\infty r \langle F'''(S)\psi^2\xi, \xi^* \rangle dr + \frac{\pi}{2} \int_0^\infty r \langle F''(S)\tilde{\zeta}_7\psi, \xi^* \rangle dr + \frac{\pi}{2} \int_0^\infty r \langle F''(S)(\tilde{\zeta}_1 + \tilde{\zeta}_2)\xi, \xi^* \rangle dr.
 \end{aligned} \tag{B7}$$

Similarly, we have

$$\begin{aligned}
 & \left\langle \frac{1}{2} F'''(S)\psi_1^2\xi_1 + F''(S)\psi_1\zeta_7 + F''(S)\xi_1\zeta_1, \xi_1^* \right\rangle_{L^2} \\
 &= \left\langle \frac{1}{2} F'''(S)\psi_1^2\xi_2 + F''(S)\psi_1\zeta_9 + F''(S)\xi_2\zeta_1, \xi_2^* \right\rangle_{L^2} \\
 &= \left\langle \frac{1}{2} F'''(S)\psi_2^2\xi_2 + F''(S)\psi_2\zeta_8 + F''(S)\xi_2\zeta_2, \xi_2^* \right\rangle_{L^2} \\
 &= \left\langle \frac{1}{2} F'''(S)\psi_2^2\xi_1 + F''(S)\psi_2\zeta_{10} + F''(S)\xi_1\zeta_2, \xi_1^* \right\rangle_{L^2} \\
 &\equiv \pi N_2'.
 \end{aligned} \tag{B8}$$

The last two terms of N_2 are obtained as

$$\begin{aligned}
 \langle \zeta_{7x_1}, \xi_1^* \rangle_{L^2} &= \int_0^{2\pi} \cos^2 \theta \cos^2 2\theta d\theta \int_0^\infty r \langle \tilde{\zeta}_{7r}, \xi^* \rangle dr + \int_0^{2\pi} \sin \theta \cos \theta \sin 2\theta \cos 2\theta d\theta \int_0^\infty r \langle \tilde{\zeta}_{8r}, \xi^* \rangle dr \\
 &+ \int_0^{2\pi} \sin^2 \theta \cos^2 2\theta d\theta \int_0^\infty \langle \tilde{\zeta}_{7r}, \xi^* \rangle dr + 2 \int_0^{2\pi} \sin \theta \cos \theta \sin 2\theta \cos 2\theta d\theta \int_0^\infty \langle \tilde{\zeta}_{7r}, \xi^* \rangle dr \\
 &- \int_0^{2\pi} \sin \theta \cos \theta \sin 2\theta \cos 2\theta d\theta \int_0^\infty \langle \tilde{\zeta}_{8r}, \xi^* \rangle dr - 2 \int_0^{2\pi} \sin^2 \theta \cos^2 2\theta d\theta \int_0^\infty \langle \tilde{\zeta}_{8r}, \xi^* \rangle dr \\
 &= \frac{\pi}{2} \int_0^\infty r \langle \tilde{\zeta}_{7r}, \xi^* \rangle dr + \frac{\pi}{2} \int_0^\infty \langle \tilde{\zeta}_{7r}, \xi^* \rangle dr - \pi \int_0^\infty \langle \tilde{\zeta}_{8r}, \xi^* \rangle dr
 \end{aligned} \tag{B9}$$

and

$$\begin{aligned}
 \langle \psi_{1x_1}, \xi_1^* \rangle_{L^2} &= \int_0^{2\pi} \cos^2 \theta \cos 2\theta d\theta \int_0^\infty r \langle \psi_r, \xi^* \rangle dr \\
 &+ \int_0^{2\pi} \sin^2 \theta \cos 2\theta d\theta \int_0^\infty \langle \psi_r, \xi^* \rangle dr \\
 &= \frac{\pi}{2} \int_0^\infty r \left\langle \psi_r - \frac{\psi}{r}, \xi^* \right\rangle dr.
 \end{aligned} \tag{B10}$$

Similarly as before, we have

$$\langle \zeta_{7x_1}, \xi_1^* \rangle_{L^2} = \langle \zeta_{9x_1}, \xi_2^* \rangle_{L^2} = \langle \zeta_{8x_2}, \xi_2^* \rangle_{L^2} = \langle \zeta_{10x_2}, \xi_1^* \rangle_{L^2} \equiv \pi N_2'' \tag{B11}$$

and

$$\begin{aligned}
 \langle \psi_{1x_1}, \xi_1^* \rangle_{L^2} &= \langle \psi_{1x_2}, \xi_2^* \rangle_{L^2} = \langle \psi_{2x_1}, \xi_2^* \rangle_{L^2} = -\langle \psi_{2x_2}, \xi_1^* \rangle_{L^2} \\
 &\equiv \pi N_2'''.
 \end{aligned} \tag{B12}$$

Therefore, we arrive at $N_2 = N_2' + N_2'' - \beta' N_2'''$.

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