Instabilities in rapid directional solidification under weak flow

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Instabilities in rapid directional solidification under weak flow

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We examine a rapidly solidifying binary alloy under directional solidification with non-equilibrium interfacial thermodynamics viz. the segregation coefficient and the liquidus slope are speed dependent and attachment-kinetic effects are present. Both of these effects alone give rise to (steady) cellular instabilities, mode S, and a pulsatile instability, mode P. We examine how weak imposed boundary-layer flow of magnitude $|\mathcal{V}|$ affects these instabilities. For small $|\mathcal{V}|$, mode S becomes a travelling wave and the flow stabilizes (destabilizes) the interface for small (large) surface energies. For small $|\mathcal{V}|$, mode P has a critical wavenumber that shifts from zero to non-zero giving spatial structure. The flow promotes this instability and the frequencies of the complex conjugate pairs each increase (decrease) with flow for large (small) wavenumbers. These results are obtained by regular perturbation theory in powers of $\mathcal{V}$ far from the point where the neutral curves cross, but requires a modified expansion in powers of $\mathcal{V}^{1/3}$ near the crossing. A uniform composite expansion is then obtained valid for all small $|\mathcal{V}|$.

I. INTRODUCTION

Additive manufacturing (AM), or three-dimensional (3D) printing, has undergone tremendous progress throughout the past three decades and offers substantial advantages over existing manufacturing methods. The layer-by-layer production capabilities offered by AM can be used to print complex parts of various geometries while minimizing manufacturing time and material wastage. AM is currently capable of printing a considerable range of products [see, e.g., 1], including metallic parts [2], aero-engine components [3], protective coatings [4], electronics [5], natural structural materials [6], tissues [7, 8], hydrogel-based materials for implantable medical devices [9] and implants and prosthesis [10]. However, parts produced by AM are susceptible to a range of undesirable effects such as distortion, compositional changes, lack of fusion defects [1], high surface roughness, layer delamination and warping [11], and denudation [12], depending on the geometry of the molten pool, temperature distribution and thermo-physical effects.

One of these effects involves the onset of flow within the melt pool as a consequence of high temperature gradients near the heat source. The resulting sharp gradients in surface tension induce Marangoni convection within the melt pool as depicted in Fig. 1 [11, 13]. Liquid to the rear of the heat source solidifies rapidly and the microstructure of the grown solid is determined by processes at the solid-liquid interface. It is the purpose of this paper to investigate the influence of flow on the morphological stability of a rapidly solidified binary alloy. Disequilibrium effects, including solute trapping and kinetic undercooling, become significant at rapid solidification rates and we investigate their roles within a thermodynamically consistent model of directional solidification in disequilibrium.

Such a configuration enables one to examine the conditions under which to expect instabilities at the solidification front under the presence of flow, given physical estimates for the magnitude of flow induced by Marangoni convection within the melt pool in a practical physical or engineering situation, and how these instabilities change when physical or material properties are varied, as well as conditions under which all instabilities are suppressed completely. Such estimates provide a starting point that may aid in gauging the choice of physical paremeters or material properties to avoid undersirable effects in practical scenarios.

The morphological stability of a binary alloy in thermodynamic equilibrium without flow has been first examined by Mullins & Sekerka [14]. Many generalisations have been provided by Coriell & McFadden [15] and an absolute stability limit exists such that the instability disappears for large enough surface energy. The effect of boundary-layer flow on the stability of a directionally solidified binary alloy under thermodynamic equilibrium has been investigated by Forth & Wheeler [16] in the limit of large Schmidt number and large Reynolds number. Hobbs & Metzener [17] conducted a long-wavelength analysis of the problem and found that imposed boundary-layer flow favours traveling waves through a destabilizing mechanism.

When the interface is no longer at thermodynamic equlibrium, as is the case at large solidification rates, phase transitions are no longer governed by the phase diagram. Departures from equilibrium have been formulated within thermodynamically consistent generalisations by

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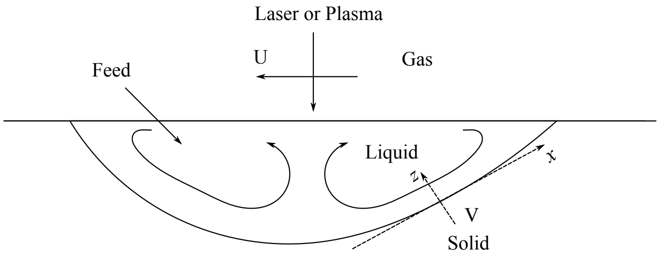

FIG. 1. Schematic of the solidification of a liquid melt pool in one form of additive manufacturing.

[18–23] to include effects that occur at high solidification rates, such as the effects of solute trapping and kinetic undercooling. These generalizations have been used in a model without flow to find that oscillatory instabilities may occur in disequilibrium, in addition to the previously observed cellular modes found at interfacial equilibrium Coriell & Sekerka [24]. The state diagram has been set out for both the cellular and the oscillatory modes by Merchant & Davis [25], and an absolute stability limit involving attachment kinetics has been found for the oscillatory mode.

We investigate the influence of boundary-layer flow, indicated in Fig. 1, on the stability of the liquid-solid interface in directional solidification in a rapid solidification environment by performing an asymptotic analysis for weak flow. In contrast to the scenario at interfacial equilibrium, two branches of instability (steady S and periodic P modes) are present here and a singular perturbation problem arises near the crossing point. We use matched asymptotic expansions to analyze perturbations to both branches and find that there is a symmetry breaking singularity where the two branches couple.

We begin by setting out the theoretical framework and formulating the linear stability theory in Sec. II. The stability of the interface under no flow is discussed in Sec. III. We investigate the effect of flow by performing regular asymptotic analyses for the steady and oscillatory branches in Sec. IV, followed by a singular perturbation analysis near the singular point in Sec. IV B. We form a uniformly-valid composite solution in Sec. IV C and revert to physical scalings in Sec. IV D. We finish with a discussion of the results and concluding remarks in Sec. V.

II. FORMULATION

We consider the rapid directional solidification of a binary alloy of local solute concentration C_l in the liquid, C_s in the solid, pulled at speed V in the negative z-direction through heat exchangers located at $\tilde{z} = \pm\tilde{L}$. For simplicity, it is assumed that $\tilde{L} \to \infty$, and the position of the resulting solid-liquid interface is given by $\tilde{z} = \tilde{h}$ in the local coordinate system of Fig. 1.

In the non-equilibrium model set forth in [18, 21–23], departure from equilibrium gives rise to a non-equilibrium local interfacial temperature T_I and local solute concentration C_s in the solid. Both of these are deviations from the equilibrium phase diagram. The local solute concentration in the solid is related to that of the liquid through

$$\tilde{C}_s = \tilde{C}_l \tilde{k}(\tilde{V}_n), \tag{1}$$

where k is the non-equilibrium segregation coefficient, which depends on the local interface speed V_n. The segregation coefficient is close to the equilibrium value k_E at low solidification rates and approaches unity at rates high enough that solute is completely trapped into the solid. A model proposed by Jackson *et al.* [19] and Aziz [20] describes this variation by

$$\tilde{k}(\tilde{V}_n) = \frac{k_E + \beta_0 \tilde{V}_n}{1 + \beta_0 \tilde{V}_n}, \tag{2}$$

where β_0 is a constant. Dimensional variables are denoted by tildes.

The arguments of Boettinger & Perepezko [21] and Boettinger & Coriell [22] for a planar interface, modified by the Gibbs-Thomson effect for curved interfaces, yield that the response function for interfacial temperature, including the effects of capillary undercooling and kinetic undercooling, is given by

$$\tilde{T}_I = T_M \left(1 + 2\tilde{H}\frac{\gamma}{L_\nu}\right) + m(\tilde{V}_n)C_l - \frac{m_E}{k_E - 1}\frac{\tilde{V}_n}{V_0}, \tag{3}$$

[see, e.g., 25, 26] where T_M is the equilibrium melting temperature of the pure material, H is the mean curvature, γ is the surface energy, L_ν is the latent heat per unit volume, m_E is the equilibrium slope of the liquidus, k_E is the equilibrium-segregation coefficient, V_0 is the upper bound for the rate at which crystallization can occur, and the change in liquidus slope due to non-equilibrium segregation is given by

$$\tilde{m}(\tilde{V}_n) = m_E \left\{ 1 - \frac{1}{k_E - 1}\left(k_E - k(\tilde{V}_n) \cdot \cdot \left[1 - \ln \frac{k(\tilde{V}_n)}{k_E} \right] \right) \right\}. \tag{4}$$

For a non-planar, three-dimensional surface,

$$2\tilde{H} = \frac{\tilde{h}_{\tilde{x}\tilde{x}}\left(1 + \tilde{h}_{\tilde{y}}^2\right) - 2\tilde{h}_{\tilde{x}}\tilde{h}_{\tilde{y}}\tilde{h}_{\tilde{x}\tilde{y}} + \tilde{h}_{\tilde{y}\tilde{y}}\left(1 + \tilde{h}_{\tilde{x}}^2\right)}{\left(1 + \tilde{h}_{\tilde{y}}^2 + \tilde{h}_{\tilde{x}}^2\right)^{3/2}}. \tag{5}$$

The solidification rate for non-planar growth is modified to

$$\tilde{V}_n = \frac{V + \tilde{h}_{\tilde{t}}}{\left(1 + |\tilde{\nabla}\tilde{h}|^2\right)^{1/2}}. \tag{6}$$

A. Basic state and non-dimensionalisation

For ease of presentation, we adopt the approximations of Merchant & Davis [25] for the thermal and solutal problems, namely, that the diffusivity of solute is negligible in the solid phase, that the diffusivity of solute in the liquid phase is much smaller than the thermal diffusivities of both phases, that the thermal conductivities of both phases are equal and that latent heat production at the interface can be neglected. The latter assumptions have the advantage that they allow for the freezing of the temperature. For the liquid, we assume that it is incompressible, that the effect of gravity is negligible and that there is no change in the density of the material as it changes phase.

In a coordinate system moving with the front at speed V, conservation of temperature, solute, momentum and mass in the liquid ($\tilde{z} > \tilde{h}$) are summarised by

$$0 = \tilde{\nabla}^2 \tilde{T}_l, \tag{7}$$

$$\frac{\partial \tilde{C}_l}{\partial \tilde{t}} - V\frac{\partial \tilde{C}_l}{\partial \tilde{z}} + \tilde{\boldsymbol{u}} \cdot \nabla \tilde{C}_l = \mathcal{D}_l \tilde{\nabla}^2 \tilde{C}_l, \tag{8}$$

$$\rho_l \left(\frac{\partial \tilde{\boldsymbol{u}}}{\partial \tilde{t}} - V\frac{\partial \tilde{\boldsymbol{u}}}{\partial \tilde{z}} + \tilde{\boldsymbol{u}} \cdot \tilde{\nabla} \tilde{\boldsymbol{u}} \right) = -\tilde{\nabla}\tilde{p} + \mu \tilde{\nabla}^2 \tilde{\boldsymbol{u}}, \tag{9}$$

$$\tilde{\nabla} \cdot \tilde{\boldsymbol{u}} = \boldsymbol{0}, \tag{10}$$

For the solid ($\tilde{z} < \tilde{h}$),

$$0 = \tilde{\nabla}^2 \tilde{T}_s, \tag{11}$$

Far field boundary conditions on the flow field and solute concentration are

$$\tilde{\boldsymbol{u}} \to U_\infty \boldsymbol{e_x} \quad \text{as} \quad \tilde{z} \to \infty, \tag{12}$$

$$\tilde{C} \to C_\infty \quad \text{as} \quad \tilde{z} \to \infty. \tag{13}$$

The remaining boundary conditions at the interface involve the local solutal balance

$$(\tilde{C}_l - \tilde{C}_s)\tilde{V}_n = -\mathcal{D}_l \frac{\partial \tilde{C}_l}{\partial \tilde{n}}, \quad \tilde{z} = \tilde{h}(\tilde{x}, \tilde{y}, \tilde{t}), \tag{14}$$

where $\tilde{\boldsymbol{n}} = (-\partial\tilde{h}/\partial\tilde{x}, \partial\tilde{h}/\partial\tilde{y}, 1)/\sqrt{1 + |\tilde{\nabla}\tilde{h}|^2}$ is the unit normal vector, and the local conservation of mass and no-slip condition

$$\tilde{\boldsymbol{u}} = \boldsymbol{0}, \quad \tilde{z} = \tilde{h}(\tilde{x}, \tilde{y}, \tilde{t}). \tag{15}$$

We shall adopt spatial and temporal scalings based on solute diffusion and scale the velocity and pressure fields by the far-field flow and viscous pressure scaling so that

$$\tilde{\boldsymbol{x}} = \frac{\mathcal{D}_l}{V}\boldsymbol{x}, \quad \tilde{t} = \frac{\mathcal{D}_l}{V^2} t,$$

$$\tilde{\boldsymbol{u}} = \boldsymbol{u}/U_\infty, \quad \tilde{p} = \frac{\mu V U_\infty}{\mathcal{D}_l} p. \tag{16$a-d$}$$

We scale the temperature and solutal field by

$$T_l = \frac{\tilde{T}_l - T_0}{G\mathcal{D}_l/V}, \quad T_s = \frac{\tilde{T}_s - T_0}{G\mathcal{D}_l/V},$$

$$C_l = \frac{\tilde{C}_l - C_\infty/k_E}{C_\infty(k_E - 1)/k_E}, \tag{17$a-c$}$$

where G is the imposed temperature gradient and T_0 is a reference equilibrium freezing temperature of the substance. The dimensionless equations become

$$0 = \nabla^2 T_l, \tag{18}$$

$$\frac{\partial C_l}{\partial t} - \frac{\partial C_l}{\partial z} + \mathcal{V}\boldsymbol{u} \cdot \nabla C_l = \nabla^2 C_l, \tag{19}$$

$$\mathcal{R}\left(\frac{\partial \boldsymbol{u}}{\partial t} - \frac{\partial \boldsymbol{u}}{\partial z} + \mathcal{V}\boldsymbol{u} \cdot \nabla \boldsymbol{u} \right) = -\nabla p + \nabla^2 \boldsymbol{u}, \tag{20}$$

$$\nabla \cdot \boldsymbol{u} = \boldsymbol{0}, \tag{21}$$

for the liquid ($z < h$),

$$0 = \nabla^2 T_s, \tag{22}$$

for the solid ($z < h$),

$$\boldsymbol{u} \to \boldsymbol{e_x} \quad \text{as} \quad z \to \infty, \tag{23}$$

$$C \to 1 \quad \text{as} \quad z \to \infty. \tag{24}$$

in the far field and

$$C_s = C_l k(V_n) \tag{25}$$

$$(C_l - C_s)V_n = -(-C_{lx}h_x - C_{ly}h_y + C_{lz}) \cdot \cdot (1 + |\nabla h|^2)^{-1/2} \tag{26}$$

$$T_l = T_s = \mathcal{M}C_l - \frac{\mathcal{M}}{(1-k_E)^2}(k_E - k(V_n)) \cdot \cdot \left(1 - \ln\frac{k(V_n)}{k_E}\right)(1 + (k_E - 1)C_l) + 2H\mathcal{M}\Gamma - \mathcal{M}\mathcal{U}V_n \tag{27}$$

at the interface $z = h$, where

$$k(V_n) = \frac{k_E + \beta V_n}{1 + \beta V_n}, \tag{28}$$

$$V_n = \frac{1 + h_t}{(1 + |\nabla h|^2)^{1/2}}, \tag{29}$$

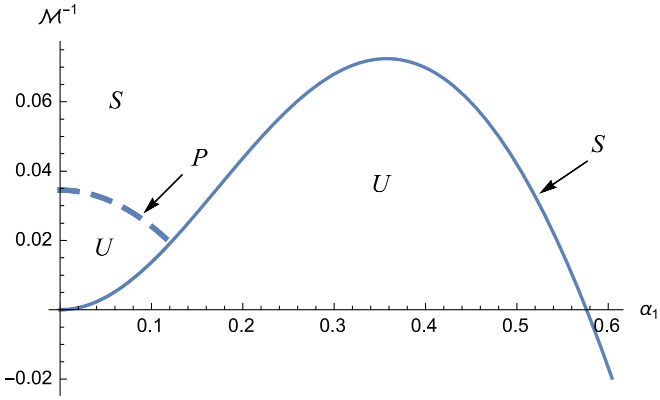

FIG. 2. The neutral stability curve under no flow. The steady branch is shown as a solid curve and the oscillatory branch is shown as a dashed curve. Parameters used: $k_E = 0.2$, $\beta = 0.1$, $\mathcal{U} = 0.01$, $\Gamma = 1$, $\mathcal{R} = 1$, $\mathcal{V} = 0$.

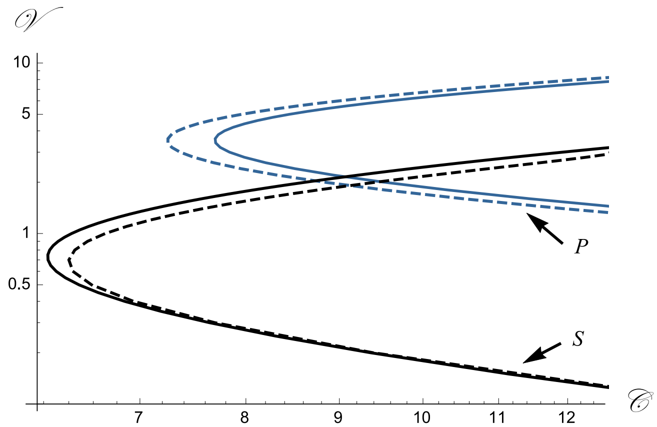

FIG. 3. Neutral stability curves in rescaled variables $(\mathscr{C}, \mathscr{V})$. Pulling speed versus far field solutal concentration for both the leading-order steady (black, solid curve) and oscillatory (blue, solid curve) branches and their perturbations due to weak flow (black, dashed and blue, dashed curves). Parameters used: $\mathscr{B} = 0.3, \mathscr{N} = 0.05, k_E = 0.8, \mathcal{R} = 1, \mathcal{V} = 1$.

and

$$2H = \frac{h_{xx}\left(1+h_y^2\right) - 2h_x h_y h_{xy} + h_{yy}\left(1+h_x^2\right)}{\left(1+h_y^2+h_x^2\right)^{3/2}}. \quad (30)$$

We note that in rapidly evolving systems, the segregation coefficient (28) – a bounded function of V_n – varies nonlinearly in V_n, which in turn varies nonlinearly, in general, with the kinetic undercooling. However, the degree of undercoolings that are commonly encountered even in rapidly evolving systems warrant a linear dependence for undercooling, (27), to be sufficient [26].

Seven dimensionless parameters appear in the above. They are the morphological number $\mathcal{M}$, the disequilibrium parameter β, the attachment-kinetics parameter $\mathcal{U}$, the equilibrium segregation coefficient k_E, which is the limiting value of the speed-dependent segregation coefficient $k(V_n)$, the dimensionless surface energy Γ, the external flow parameter $\mathcal{V}$ and the inverse Schmidt number $\mathcal{R}$. Explicit formulas for them are given by

$$\mathcal{M} = \frac{m_E(k_E-1)C_\infty V}{\mathcal{D}_l G k_E}, \quad \beta = \beta_0 V, \quad (31a,b)$$

$$\mathcal{U} = \frac{V k_E}{(k_E-1)^2 C_\infty V_0}, \quad \mathcal{V} = U_\infty / V, \quad (32a,b)$$

$$\Gamma = \frac{T_M \gamma V k_E}{L_\nu \mathcal{D}_l m_E (k_E-1) C_\infty}, \quad \mathcal{R} = \frac{\rho_l \mathcal{D}_l}{\mu}. \quad (33a,b)$$

This system possesses a steady-state solution, for which the interface is planar, given by

$$T = z + \mathcal{M}\gamma, \quad C_{l0} = 1 - \delta e^{-z}, \quad (34a,b)$$

$$h_0 = v_0 = w_0 = p_0 = 0, \quad u_0 = 1 - e^{-z\mathcal{R}}, \quad (35a,b)$$

where

$$\gamma = -\frac{k_E}{(k_E-1)^2}\ln\left(\bar{k}/k_E\right) + \frac{\beta}{(1-k_E)(\beta+k_E)} - \mathcal{U}, \quad (36)$$

$$\bar{k} = \frac{k_E+\beta}{1+\beta}, \quad \delta = \frac{k_E}{\beta+k_E}. \quad (37a,b)$$

B. Linear stability analysis

We investigate the stability of the planar-interface solution using linear perturbation theory and search for normal-mode solutions with growth rate σ and wave-vector $\boldsymbol{\alpha} = (\alpha_1, \alpha_2)$ by writing $X = X_0(z) + \epsilon \hat{X}(\boldsymbol{x}, t)$, where $\hat{X}(\boldsymbol{x}, t) = X_1(z)\exp(\sigma t + \mathrm{i}(\alpha_1 x + \alpha_2 y))$ for $X = (u, v, w, p, C_l)$, and $h = \epsilon\hat{\eta}(\boldsymbol{x}, t) = \epsilon\eta\exp(\sigma t + \mathrm{i}(\alpha_1 x + \alpha_2 y))$. The stability of the system is determined by the sign of q, where $\sigma = q + i\omega$, and $q, \omega \in \mathbb{R}$, with stability if $q < 0$ for all wavenumbers and instability otherwise. The instability is steady if $\omega = 0$ and oscillatory if $\omega \neq 0$.

Eliminating pressure and horizontal velocity yields a fourth-order differential equation,

$$\left(\sigma - \frac{\mathrm{d}}{\mathrm{d}z} + \mathrm{i}\alpha_1\mathcal{V}u_0 - \frac{1}{\mathcal{R}}\left(\frac{\mathrm{d}^2}{\mathrm{d}z^2} - \alpha_1^2 - \alpha_2^2\right)\right)\cdot$$
$$\cdot\left(\frac{\mathrm{d}^2}{\mathrm{d}z^2} - \alpha_1^2 - \alpha_2^2\right)w_1 = \mathrm{i}\alpha_1\mathcal{V}u_0' w_1, \quad (38)$$

for the vertical component of velocity, with interfacial boundary conditions

$$w_1 = 0, \quad w_1' = \mathrm{i}\alpha_1\mathcal{R}\eta u_0', \quad z = 0 \quad (39a,b)$$

and far field conditions

$$w_1 \to 0, \quad w_1' \to 0 \quad \text{as} \quad z \to \infty. \tag{40a, b}$$

Here, the prime denotes differentiation with respect to z. It suffices to consider this fourth-order system to determine the flow field and so our perturbation problem can be formulated in terms of w_1, C_{l1} and η_1 only.

The perturbed solutal field satisfies

$$\begin{aligned} C_{l1}'' - (\alpha_1^2 + \alpha_2^2) C_{l1} &= \mathcal{V} w_1 C_{l0}' + \mathrm{i}\alpha_1 \mathcal{V} u_0 C_{l1} + \\ &\quad + \sigma C_{l1} - C_{l1}', \end{aligned} \tag{41}$$

in $z > 0$ with the far-field condition

$$C_{l1} \to 0 \quad \text{as} \quad z \to \infty, \tag{42}$$

and interfacial conditions

$$\begin{aligned} -(\eta C_{l0}'' + C_{l1}') &= \left(1 - \frac{\bar{k}\beta}{(1+\beta)}\right) \sigma \eta C_{l0} \\ &\quad + (1 - \bar{k})(\eta C_{l0}' + C_{l1}), \end{aligned} \tag{43}$$

at $z = 0$, originating from a perturbation to the local solute conservation condition (26), and

$$\begin{aligned} 0 &= (k_E - 1)\Big[(\beta+1)\eta + \mathcal{M}\Big(- \eta C_{l0}' - C_{l1} + \\ &\quad + (\beta+1)\eta \left(\Gamma \left(\alpha_1^2 + \alpha_2^2\right) + \sigma \mathcal{U}\right) \Big)\Big] + \\ &\quad \mathcal{M} \ln\left(\bar{k}/k_E\right) \Big[\sigma\beta\eta\Big((1 - k_E) C_{l0} - 1\Big) + \\ &\quad + (\beta+1)^2 (\eta C_{l0}' + C_{l1}) \bar{k}\Big] / (\beta + 1), \end{aligned} \tag{44}$$

at $z = 0$, originating from a perturbation to the interfacial temperature condition (27). The latter condition determines the position of the perturbed interface. This system of perturbation equations is a differential eigenvalue problem for (C_{l1}, w_1, η), which admits nontrivial solutions only for certain eigenvalues σ.

Setting $\mathcal{V} = 0$ turns off the external flow and recovers the result of Merchant & Davis [25]. In this case, the characteristic equation can be obtained in closed form. We display plots of relevant results for comparison in Sec. III.

III. ZERO FLOW

In the absence of flow, the linear stability problem gives rise to the following characteristic equation

$$\begin{aligned} \mathcal{M}^{-1} = m_0 &\equiv \frac{\Gamma_s}{\beta+1}\Bigg((\beta + k_E) - \beta\sigma + \\ &\quad - \frac{2\,(\beta + k_E)\,(\beta + k_E + \sigma)}{(\beta+1)\lambda_1 + \beta + 2k_E - 1}\Bigg) \end{aligned}$$

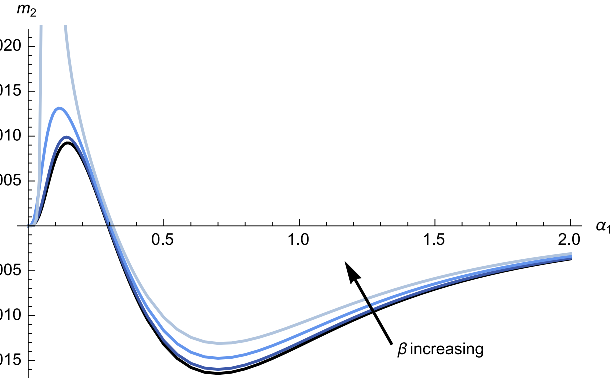

FIG. 4. Values of the second-order contributions m_2 for the steady mode for varying disequilibrium parameters $\beta = 0.01, 0.02, 0.05, 0.1$. Parameters used: $k_E = 0.5, \mathcal{U} = 0.1, \Gamma = 1, \mathcal{R} = 1$.

$$+ \sigma(\mathcal{U}_s - \mathcal{U}) - \alpha^2 \Gamma, \tag{45}$$

obtained by Merchant & Davis [25], where

$$\lambda_1 = \sqrt{4\alpha^2 + 4\sigma + 1}, \tag{46}$$

$$\Gamma_s = \frac{k_E \left((1 - k_E) + (\beta + k_E) \log\left(\frac{\beta + k_E}{\beta k_E + k_E}\right)\right)}{(1 - k_E)\,(\beta + k_E)^{\,2}}, \tag{47}$$

and

$$\mathcal{U}_s = \frac{\beta k_E}{(\beta+1)\,(\beta + k_E)^{\,2}}. \tag{48}$$

Γ_s and $\mathcal{U}_s$ are the critical values of Γ and $\mathcal{U}$, respectively, beyond which all disturbances are stabilized under no flow.

At marginal stability, we have $\sigma = i\omega_0$, $\omega_0 \in \mathbb{R}$ (we reserve the subscript 0 to denote quantities involving no flow). There are two possible scenarios. The first is one in which $\omega_0 = 0$, which corresponds to the steady instability giving mode S. The second is one in which $\omega_0 \neq 0$, which corresponds to the oscillatory instability giving mode P. The values of ω_0 are obtainable by requiring that $\Im(m_0) = 0$, which we expect on physical grounds. The values of ω_0 can be seen graphically against α_1 for the mode P in Fig. 8. The neutral curves for the S and P branches, as well as the regions of instability, are depicted in Fig. 2. The neutral curve for the P instability consists of two branches with complex conjugate frequencies.

By non-dimensionalizing with respect to capillary scales rather than diffusion scales, as in [25], it is possible to isolate the dimensionless pulling speed and far field solute concentration as independently controllable parameters. Such a rescaling is of benefit from the experimental perspective, where these two parameters, specifically, are controllable. To scale length and time against the capillary scales $l \sim (\gamma T_M/(L_\nu G)^{1/2}$, $t \sim \gamma T_M/(L_\nu G D_l)$, we

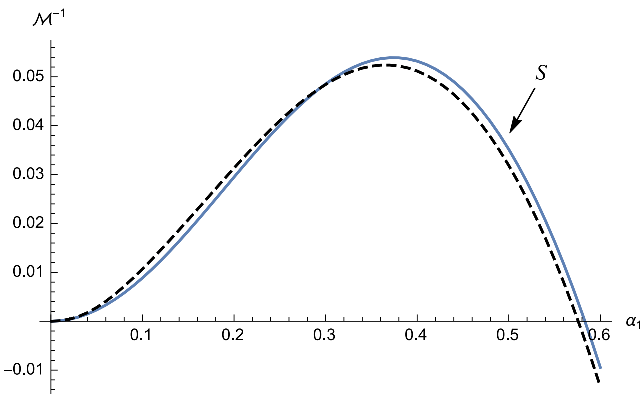

FIG. 5. Plots of m_0 (solid) and $m_0 + \mathcal{V}^2 m_2$ (dashed) against α_1 for mode S, where $\beta = 0.01$, $k_E = 0.5$, $\mathcal{U} = 0.1$, $\Gamma = 1$, $\mathcal{R} = 1$, $\mathcal{V} = 0.5$. Only the steady branch appears at leading-order.

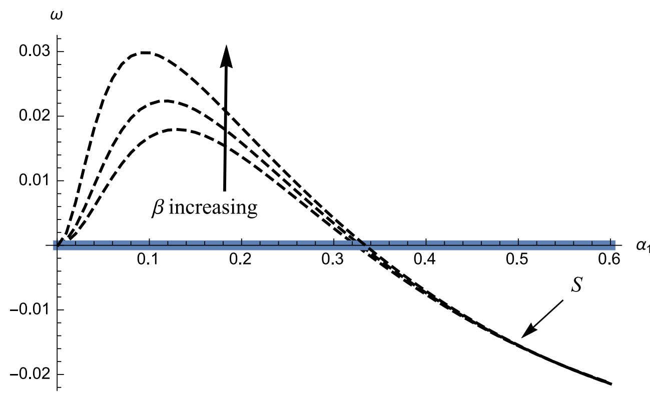

FIG. 6. Frequencies ω_0 (solid, thick) and $\omega_0 + \mathcal{V}\omega_1$ (dashed) against α_1 for mode S for various disequilibrium parameters $\beta = 0, 0.025, 0.05$. Parameters used: $k_E = 0.5$, $\mathcal{U} = 0.1$, $\Gamma = 1$, $\mathcal{R} = 1$.

use the following rescaling specified by Merchant & Davis [25]:

$$\mathcal{M} = \mathscr{V}\mathscr{C}, \qquad \Gamma = \mathscr{V}/\mathscr{C}, \quad \beta = \mathscr{B}\mathscr{V}, \qquad (48\,a-c)$$

$$\mathcal{U} = \mathscr{V}\mathscr{N}/\mathscr{C}, \quad \boldsymbol{\alpha} = \mathscr{A}/\mathscr{V}, \quad \sigma = \mathscr{S}/\mathscr{V}^2. \qquad (49\,a-c)$$

The resulting neutral stability curves can be obtained implicitly and are shown in Fig. 3. The marginal dimensionless pulling speed versus the concentration is displayed for both the leading-order steady and oscillatory branches.

The following behaviour has been reported by Merchant & Davis [25] absent flow. When $\mathscr{C}$ is fixed, say $\mathscr{C} = 8$, and $\mathscr{V}$ increases from 0 to a critical value $\mathscr{V} \approx 0.3$, the solid-liquid interface remains planar. Steady cells appear as $\mathscr{V}$ increases past its critical value. The cells deepen and their wavelength decreases as $\mathscr{V}$ is increased further, until a cellular-dendritic transition, not explained by linear theory, occurs. As $\mathscr{V}$ increases further, the cells return and then finally disappear at a critical $\mathscr{V} \approx 2$. The solid-liquid interface becomes planar within a window of stability. As $\mathscr{V}$ increases further past a critical value $\mathscr{V} \approx 3$, the pulsatile mode becomes unstable, producing solute bands periodic in the direction of solidification. These disappear and the interface regains stability once $\mathscr{V}$ reaches a critical value of $\mathscr{V} \approx 5$. Nonlinear effects become important for intermediate values of $\mathscr{V}$ within this range. For higher values of $\mathscr{C}$, both steady and pulsatile modes of instability co-exist within a region of intersection.

IV. ASYMPTOTICS FOR WEAK FLOW

In this section, we analyze the asymptotic behaviour of the solid-liquid interface for both branches for weak flows $\mathcal{V} \ll 1$. For clarity, we present only the results of our analysis in this section, and refer the reader to the Appendices for detailed derivations.

A singular perturbation problem arises in scenarios in which both steady and oscillatory branches appear, as a result of the occurance of the singular point on the neutral curve, at which the two oscillatory branches meet the steady branch giving rise to a multiple eigenvalue. We treat this by developing matched asymptotic expansions involving regular perturbations in an outer region, away from the singular point, and singular perturbations in an inner region, close to the singular point.

A. Regular perturbations

We proceed by expanding in the external-flow-parameter $\mathcal{V}$ as follows

$$C_{l1} = C_{l10} + \mathcal{V}C_{l11} + \mathcal{V}^2 C_{l12} + \cdots, \qquad (50)$$

$$w_1 = w_{10} + \mathcal{V}w_{11} + \mathcal{V}^2 w_{12} + \cdots. \qquad (51)$$

We are interested in marginal stability, which occurs when $q = 0$. The value of $\mathcal{M}$ for which this occurs can be expanded in $\mathcal{V}$ as

$$\mathcal{M}^{-1} = m_0 + \mathcal{V}m_1 + \mathcal{V}^2 m_2 + \cdots. \qquad (52)$$

The resulting instability may have either $\omega_0 = 0$ or $\omega_0 \neq 0$ and we write

$$\omega = \omega_0 + \mathcal{V}\omega_1 + \mathcal{V}\omega_2 + \cdots. \qquad (53)$$

At zeroth-order ($\mathcal{V} = 0$), there are situations in which only mode S appears, situations in which only mode P appears, and situations in which both modes S and P appear.

We find that the coefficient m_1 in (52) vanishes for mode S. As expected on physical grounds, the direction of flow, given by the sign of $\mathcal{V}$, should not influence the onset of the leading-order steady branch of the instability.

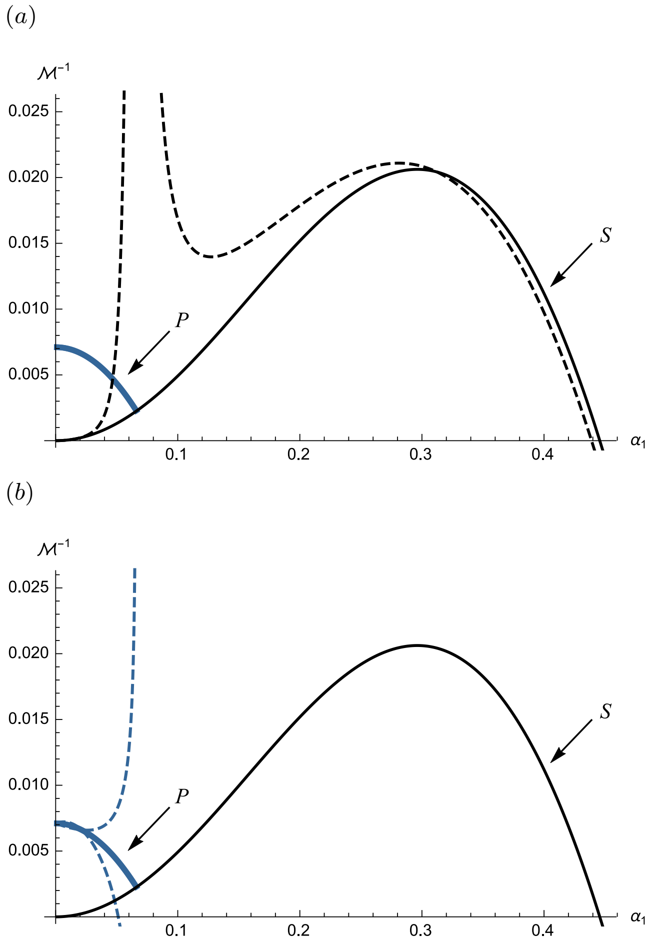

FIG. 7. Perturbations to the neutral curves for the leading-order (a) steady (dashed, black) and (b) oscillatory (dashed, blue) branches. The unperturbed neutral curve is overlaid for the steady (solid, black) and oscillatory (solid, blue) branches. Both the steady and the oscillatory branches appear at leading-order. There are two oscillatory branches and they lead to distinct inverse morphological numbers at higher order. Parameters used: $\beta = 0.1$ $k_E = 0.5$, $\mathcal{U} = 0.1$, $\Gamma = 1$, $\mathcal{R} = 1$, $\mathcal{V} = 0.5$.

The influence of flow on mode S can be determined by examining second-order corrections in $\mathcal{V}$.

Despite $m_1 = 0$ for mode S, there is a nonzero contribution $\omega_1 \neq 0$. The frequency ω_1 is shown in Fig. 6 in a situation in which only the mode S appears, and in Fig. 8 in a situation in which both modes S and P appear.

Thus, for S,

$$\mathcal{M}^{-1} = m_0 + \mathcal{V}^2 m_2 + \cdots , \tag{54}$$

and for P,

$$\mathcal{M}^{-1} = m_0 + \mathcal{V} m_1 + \cdots . \tag{55}$$

Following the details presented in Appendix A, we find

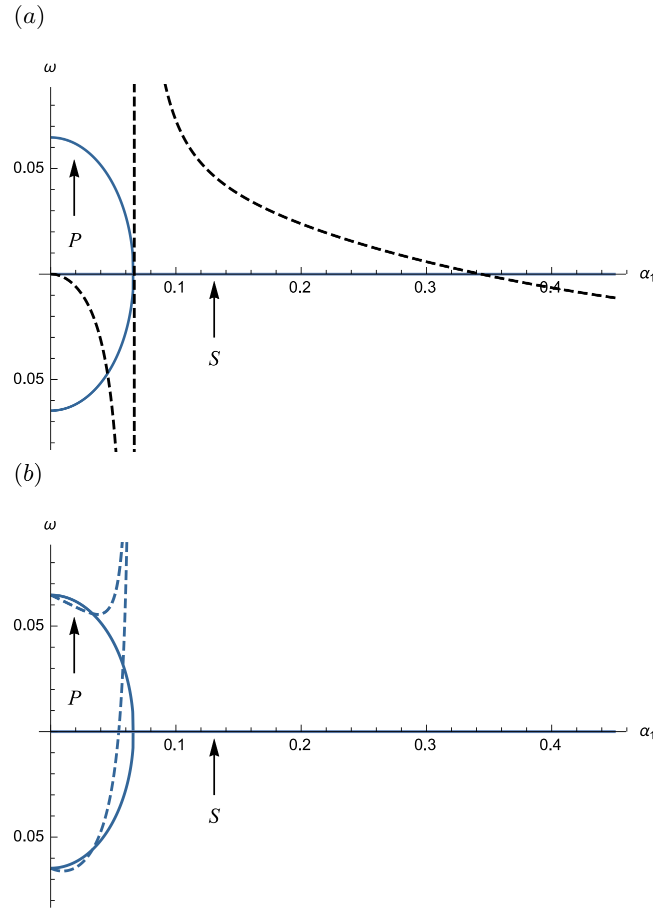

FIG. 8. (a) Perturbations to the frequencies for the leading-order (a) steady (dashed, black) and (b) oscillatory (dashed, blue) branches. The unperturbed frequencies ω_0 are overlaid for the steady (solid, black) and oscillatory (solid, blue) branches. Parameters used: $\beta = 0.1$ $k_E = 0.5$, $\mathcal{U} = 0.1$, $\Gamma = 1$, $\mathcal{R} = 1$, $\mathcal{V} = 0.5$.

for mode P, $\omega_0 \neq 0$ that

$$m_1 = \frac{\mathcal{L}\,(\beta + k_E) + (1 - k_E)}{\eta(\beta+1)\,(1-k_E)} \sum_{j=1}^{4} \Re(P_{1j}), \tag{56}$$

as given in (A35), where the P_{1j} are specified in Appendix A, and for mode S, $\omega_0 = 0$,

$$m_2 = \frac{\mathcal{L}\,(\beta + k_E) + (1 - k_E)}{\eta(\beta+1)\,(1-k_E)} \sum_{j=1}^{7} \Re(J_{21j}), \tag{57}$$

as given in (A47), where the constants J_{21j} are given in Appendix C.

The perturbations m_2 for mode S are diplayed in Fig. 4, and plots of the perturbations m_1 and the resulting $\mathcal{M}^{-1} = m_0 + \mathcal{V} m_1$ are shown in Fig. 7. There are two oscillatory branches at leading order, yielding complex conjugate roots $\pm i\omega_0$. These two branches correspond to a single $\mathcal{M}^{-1}$ at leading order. At higher order, these two branches split. Both of these are displayed in Fig. 7.

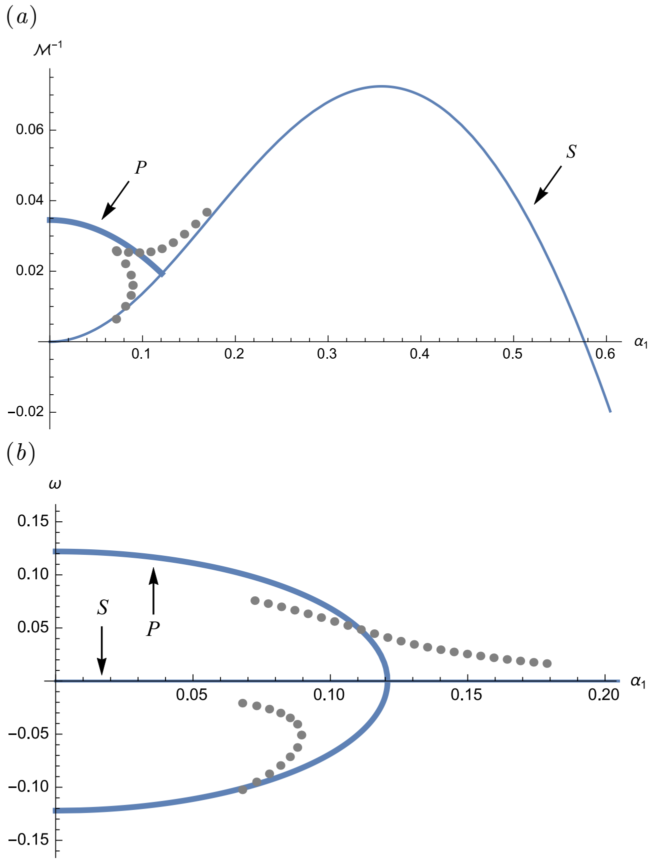

FIG. 9. Singular perturbation results (dotted curves) near the singular root for the inverse morphological number in (a) and frequency in (b) against the wavenumber. Zeroth order results (no flow) are overlain as solid curves (the steady branch is shown using thin solid curves, and the oscillatory branch is shown using thick solid curves). Parameters used: $\beta = 0.1$ $k_E = 0.5$, $\mathcal{U} = 0.1$, $\Gamma = 1$, $\mathcal{R} = 1$, $\mathcal{V} = 0.5$.

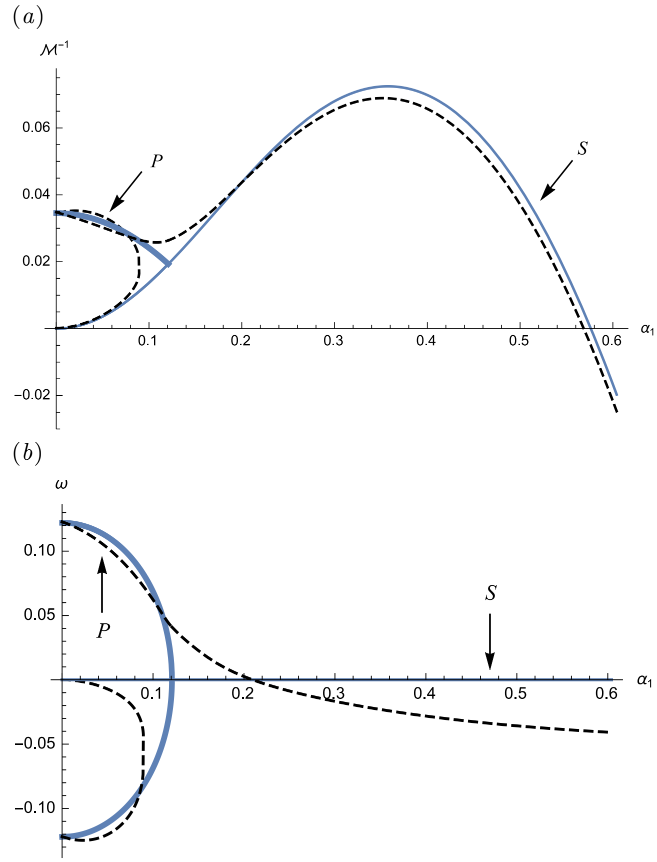

FIG. 10. The composite solution (dashed), uniformly valid for all wavenumbers, for the inverse morphological number in (a) and frequency in (b), against the wavenumber. Zeroth order results (no flow) are shown in as solid curves (the steady branch is shown using thin solid curves, and the oscillatory branch is shown using thick solid curves). Parameters used: $\beta = 0.1$ $k_E = 0.5$, $\mathcal{U} = 0.1$, $\Gamma = 1$, $\mathcal{R} = 1$, $\mathcal{V} = 0.5$.

Note that only the upper branch determines the instability boundary, however, the other branch may lead to higher growth rates and we choose to display both of the branches for this reason. The corresponding frequency perturbation ω_1 and $\omega_0 + \mathcal{V}\omega_1$ are shown in Fig. 8 for both the leading-order oscillatory and steady branches in the scenario in which both of them appear at leading-order.

Close to the crossing point, the regular perturbation expansions diverge as seen in Figs. 7 and 8. This signals a need for a singular perturbation analysis near this point, which we perform in Sec. IV B.

B. Singular perturbations

Of interest for the current section is the point at which the two branches meet. This is a junction of three branches: S and P (complex conjugates). We have seen in the previous section that a regular perturbation expansion fails in the vicinity of this singular root. In order to resolve for the behaviour near this multiple root, we pose an asymptotic expansion in the parameter $\mathcal{V}^{1/3}$, of the form

$$w_1 = \tilde{w}_{10} + \mathcal{V}^{1/3}\tilde{w}_{10} + \mathcal{V}^{2/3}\tilde{w}_{12} + \cdots , \tag{58}$$

$$C_{l1} = \tilde{C}_{l10} + \mathcal{V}^{1/3}\tilde{C}_{l10} + \mathcal{V}^{2/3}\tilde{C}_{l12} + \cdots . \tag{59}$$

We are again interested in marginal stability, which occurs when $q = 0$ and this time expand the value of $\mathcal{M}^{-1}$ for which this occurs as

$$\mathcal{M}^{-1} = \tilde{m}_0 + \mathcal{V}^{2/3}\tilde{m}_2 + \mathcal{V}^{4/3}\tilde{m}_4 + \cdots , \tag{60}$$

and the imaginary part of σ as

$$\omega = \mathcal{V}^{1/3}\tilde{\omega}_1 + \mathcal{V}\tilde{\omega}_3 + \mathcal{V}^{5/3}\tilde{\omega}_5 + \cdots . \tag{61}$$

Taking $\mathcal{V} \to 0$ leads to $\sigma = 0$, which is the value of σ at the singular root, as expected. We seek an inner expansion near the singular root, which occurs at some wavenumber $\alpha_1 = \alpha_{10}$. The appropriate inner scaling for the wavenumber is

$$\alpha_1 = \alpha_{10} + \alpha_{11}\mathcal{V}^{2/3}, \tag{62}$$

which follows from the square-root dependence of the two oscillatory frequencies on the wavenumber in the vicinity of the singular root at leading-order.

It is necessary to consider expansions up to third-order in $\mathcal{V}^{1/3}$ to obtain leading-order information on the influence of flow. We lay out our analysis order by order, in Appendix B.

We find, at first order in $\mathcal{V}^{1/3}$, that the wavenumber α_{10} satisfies

$$\begin{aligned}\frac{\beta\Gamma_s}{\beta+1}+(\mathcal{U}-\mathcal{U}_s) \;&= \frac{4\Gamma_s\,(\beta+k_E)}{(\beta+1)^2\sqrt{4\alpha_{10}^2+1}}\cdot\\ &\cdot\left(\frac{\Omega_1(\alpha_{10})}{\left[\Omega_2(\alpha_{10})\right]^2}-\frac{1}{4}\right),\end{aligned} \tag{63}$$

where

$$\Omega_1(\alpha_{10})=(\beta+2)\beta^2+(\beta+3)\beta k_E-(\beta+1)^2\alpha_{10}^2+k_E^2, \tag{64}$$

and

$$\Omega_2(\alpha_{10})=\beta+2k_E-1+(\beta+1)\sqrt{4\alpha_{10}^2+1}. \tag{65}$$

This relation ensures that $\alpha_1=\alpha_{10}$ corresponds to the multiple root. Using it, one may obtain an asymptotic expression for α_{10} in the form

$$\begin{aligned}\alpha_{10} \;=\; &\frac{\sqrt{k_E}}{\sqrt{2k_E+1}}\beta^{1/2}+\\ &-\frac{\left(2k_E^3+7k_E^2-1\right)}{2\sqrt{k_E}\,(2k_E+1)^{5/2}}\beta^{3/2}+\mathcal{O}(\beta^2),\end{aligned} \tag{66}$$

for β small in the limit $\mathcal{U}=0$. We find, at second order in $\mathcal{V}^{1/3}$, that $\tilde{m}_2$ satisfies

$$\begin{aligned}\tilde{m}_2 = &-2\Gamma\alpha_{10}\alpha_{11}+\\ &-\frac{(\mathcal{L}(k_E+\beta)+1-k_E)}{(\beta+1)\,(k_E-1)\,\eta}\Re\left(\tilde{J}_{2,1,1}\right),\end{aligned} \tag{67}$$

which depends on $\tilde{\omega}_1$, and $\tilde{J}_{2,1,1}$ are constants that are given in Appendix C. We determine $\tilde{\omega}_1$ by examining the third order in $\mathcal{V}^{1/3}$ and find that $\tilde{\omega}_1$ solves the cubic

$$\gamma_1\tilde{\omega}_1^3+\gamma_2\alpha_{11}\tilde{\omega}_1+\gamma_3=0, \tag{68}$$

where $\gamma_1,\ldots,\gamma_3$ are given in Appendix B. With $\tilde{\omega}_1$ known, $\tilde{m}_2$ is determined from (67).

The resulting singular perturbations for the inverse morphological number and frequency near the singular root are shown in Fig. 9. The topology of the neutral curves changes near the singular root. The low wavenumber steady branch joins with one of the oscillatory branches and the high-wavenumber steady branch joins with the other oscillatory branch. The regions of stability and instability become disjoint.

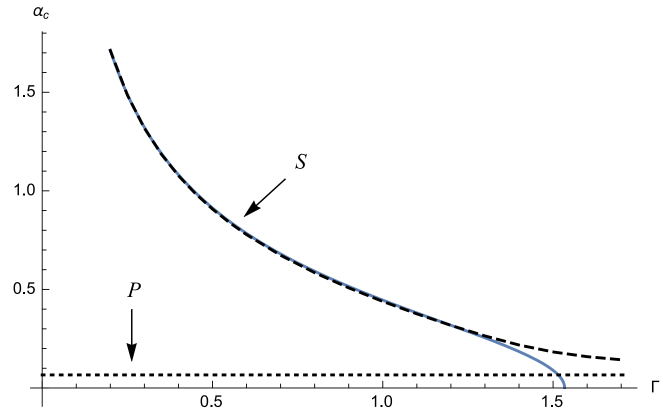

FIG. 11. Cut-off wavenumber α_c for the onset of instability with flow (dashed) and without flow for mode S (solid) and mode P (dotted). Parameters used: $k_E=0.5$, $\beta=0.1$, $\mathcal{U}=0.1$, $\mathcal{R}=1$, $\mathcal{V}=0.5$.

C. Composite solution

As the contributions of flow to the leading-order oscillatory branch appear at $\mathcal{O}(\mathcal{V})$, the inner solution (67) is sufficient to determine a uniformly valid solution for the leading-order oscillatory branch. However, this is not the case for the leading-order steady branch, where the leading-order effects of flow appear at $\mathcal{O}(\mathcal{V}^2)$. It is therefore necessary to derive higher-order terms in the singular expansion for the inverse morphological number in this region.

As shown in Appendix B, the form of $\tilde{m}_4$ is given by

$$\tilde{m}_4=\rho_1\tilde{\omega}_1^4+\rho_2\alpha_{11}\tilde{\omega}_1^2+\rho_3\tilde{\omega}_1+\rho_4\alpha_{11}^2+\rho_5\tilde{\omega}_3\tilde{\omega}_1, \tag{69}$$

where $\rho_1,\ldots,\rho_5$ are numerical constants that depend on α_{10} and the physical parameters. This expression for $\tilde{m}_4$ depends on $\tilde{\omega}_3$, which we determine in Appendix B by considering the next highest order and obtain the equation

$$\begin{aligned}(\rho_6\tilde{\omega}_1^2+\rho_7\alpha_{11})\tilde{\omega}_3+\rho_8\tilde{\omega}_1^5+\rho_9\alpha_{11}\tilde{\omega}_1^3+&\\ +\rho_{10}\tilde{\omega}_1^2+\rho_{11}\alpha_{11}^2\tilde{\omega}_1+\rho_{12}\alpha_{11}=0,&\end{aligned} \tag{70}$$

for $\tilde{\omega}_3$ by requiring the morphological number to be real, giving that $\tilde{m}_5=0$, and noting that $\alpha_1=\alpha_{10}$ corresponds to the singular point that eliminates $\tilde{\omega}_5$. Here, $\rho_6,\ldots,\rho_{12}$ are numerical constants that depend on the parameters. Given the form of $\tilde{\omega}_3$ from (70), $\tilde{m}_4$ is fully determined via (69). This determines the inverse morphological number to the required order in the inner region for a smooth composite solution to be formed.

An additive composite solution is formed for $\mathcal{M}^{-1}$ and ω by matching the inner solutions (60) and (61) with the leading-order outer solutions $\mathcal{M}^{-1}\sim m_0+\mathcal{V}^2m_2$, $\sigma\sim\mathcal{V}\omega_1$, for the steady branch, and $\mathcal{M}^{-1}\sim m_0+\mathcal{V}m_1$, $\sigma\sim\omega_0+\mathcal{V}\omega_1$, for the oscillatory branch. Fig. 10 shows the

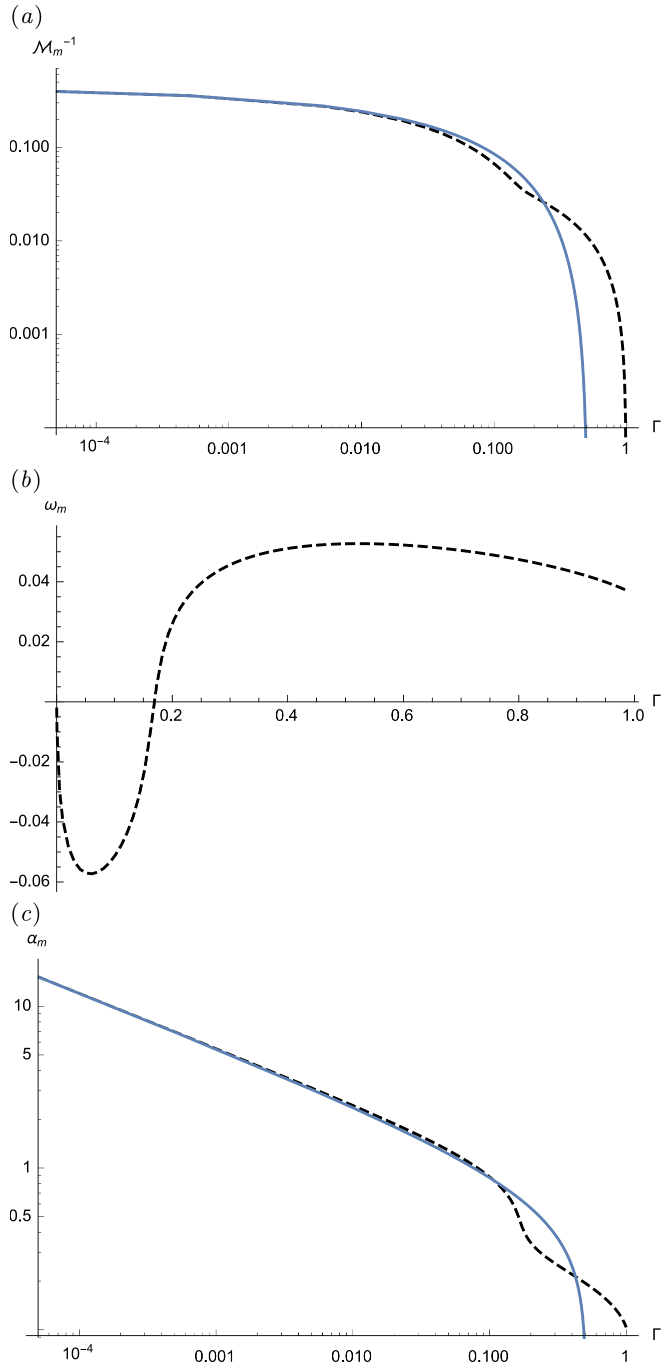

FIG. 12. (a) Maximal $\mathcal{M}_m^{-1}$ for mode S and the associated (b) frequency ω_m and (c) wavenumber α_m as a function of Γ with flow (dashed) and without flow (solid) for $k_E = 0.7$, $\beta = 1$, $\mathcal{U} = 0.2$, $\mathcal{R} = 1$.

composite solution against the results without flow. The singularity at the junction of the three branches is well resolved and the solution is uniformly valid throughout the domain.

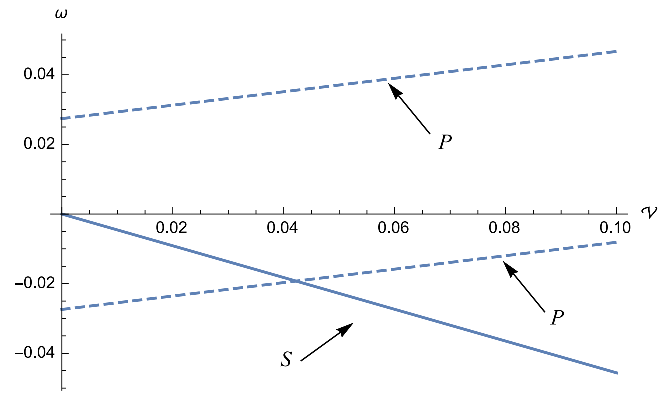

FIG. 13. Frequency $\omega_0 + \mathcal{V}\omega_1$ as a function of $\mathcal{V}$ for the steady (solid) and two oscillatory (dashed) branches for $\alpha = 0.06$, $k_E = 0.5$, $\beta = 0.1$, $\mathcal{U} = 0.1$, $\Gamma = 1$, $\mathcal{R} = 1$.

D. Physical Scalings

We revert back to the natural scalings $(48\,a - c)$ and $(49\,a - c)$, which use surface energy to scale the quantities, and discuss the effect of flow on the resulting neutral curves. An advantage of the natural scalings is that they give neutral curves that appear similar to the dimensional ones.

We obtain the neutral stability curves implicitly via the transformations $(48\,a - c)$–$(49\,a - c)$ and display them graphically in Fig. 3. The neutral curves without the presence of flow are overlain in the figure, for comparison. Displayed are the marginal dimensionless pulling speed versus the concentration for both the leading-order steady and oscillatory branches. The presence of flow enlarges the region of instability in favour of the leading-order oscillatory branch. The minimal value of $\mathscr{C}$ for which oscillatory instabilities occur is smaller once perturbed by the presence of flow. On the other hand, the range of values of $\mathscr{C}$ for which leading-order steady instabilities occur reduces under the presence of flow. In this way, the presence of flow eliminates instabilities for low enough concentrations.

V. DISCUSSION AND CONCLUSIONS

We have analyzed the effects of weak flow on the directional solidification of a binary alloy with an interface that departs from thermodynamic equilibrium. In particular, with no flow the linearized instabilities of the front display two modes [25]. On their neutral curves there is a steady, cellular mode S and a time-periodic mode P, and we study how these are changed by the imposition of a weak shear flow of boundary layer type. Perturbation methods are used for flow magnitude $|\mathcal{V}| \ll 1$. The mode S is steady for $\mathcal{V} = 0$ with maximizing wavenumber α_m, and cut-off wavenumber α_c of unit order. In Fig.

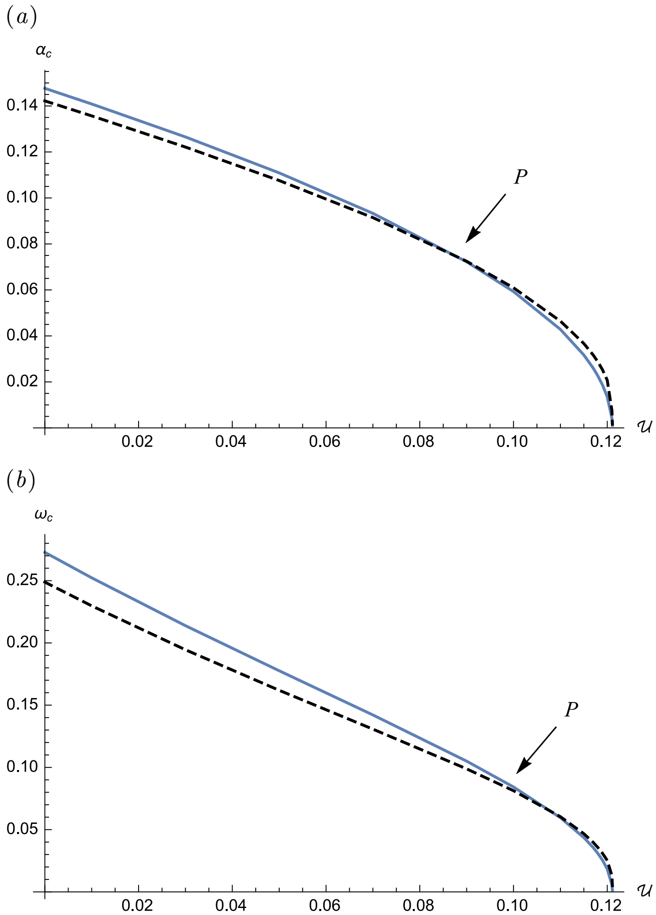

FIG. 14. (a) Cut-off wavenumber α_c and (b) associated frequency ω_c for mode P as a function of $\mathcal{U}$ with (dashed) and without (solid) flow for $k_E = 0.7$, $\beta = 1$, $\Gamma = 2$, $\mathcal{R} = 1$.

2, $\alpha_m \approx 0.35$ and $\alpha_c \approx 0.58$, and the angular frequency $\omega = 0$. The critical morphological number is given by $\mathcal{M}_c^{-1} \approx 0.074$. The neutral curve in natural coodinates is also shown in Fig. 3.

The imposition of flow delays the instability as seen in Figs. 3 and 5. The instability exists for $\Gamma < \Gamma_s$, the absolute stability boundary, and Γ_s increases with increasing $\mathcal{V}$. Both α_m and α_c decrease with $\mathcal{V}$ for moderate Γ, as seen in Fig. 5. The previously steady mode now travels due to $\mathcal{V}$ as shown in Fig. 13, where ω is linear in $\mathcal{V}$ and negative for small α and the maximizing ω_m is negative for small Γ as seen in Fig. 12b, meaning that the wave travels into the shear consistent with dendrites growing into oncoming flow, an effect of the concentration of solute being asymmetric fore and aft (Jeong *et al.* [27]). Here, $\omega > 0$ for large α and $\omega_m > 0$ for large Γ as seen in Fig. 12b.

The mode P has $\alpha_m = 0$ for $\mathcal{V} = 0$ as shown in Fig. 2, so that the mode is pulsatile with no spatial structure, but $\alpha_m \neq 0$ when $\mathcal{V} \neq 0$ as seen in Fig. 10a, where $\alpha_m \approx 0.03$. The mode appears as a complex conjugate pair for $\mathcal{V} \neq 0$ but each frequency increases with flow for large α as shown in Fig. 13, the corrections again linear in $\mathcal{V}$. The frequencies decrease with flow for small α as seen in Fig. 10b. The mode exists for kinetics parameter $\mathcal{U} < \mathcal{U}_s$, a second absolute stability boundary. Flow promotes instability P as shown in Fig. 3.

The cutoff wavenumber for the onset of instability smoothly transitions from one branch of instability to the other as seen in Fig. 11. Fig. 3 illustrates the effect of flow on modes S and P together. Mode P is destabilized and mode S is stabilized at the nose of the neutral stability curve. Waves appear for both modes in the presence of flow. Experiments [28–31] on metallic systems in rapid solidification display bands, microstructures that have alternate layers of dendrites/cells and structure-free material periodic in the pulling direction. Merchant and Davis [25] show that these bands are always located in the sector of $(\mathscr{C}, \mathscr{V})$ space, see Fig. 3, common to both the S and P instabilities. With flow, this sector moves downwards and to the left (right) for high (low) values of the naturally scaled attachment kinetics parameter $\mathscr{N}$, (3.5 a), meaning that banding would occur at lower pulling speeds and lower (higher) concentrations than without flow. Even more importantly, with flow the mode S, dendrites/cells, now travel, and the mode P now has a non-zero critical wave number inducing spatial structure in the P mode. Thus, banding morphologies are intrinsically altered by the presence of flow.

ACKNOWLEDGMENTS

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Appendix A: Regular perturbations

1. Zeroth order

The zeroth-order perturbed vertical velocity and solutal fields satisfy

$$\alpha^2 w_{10}(z)\left(\alpha^2 + i\omega_0\mathcal{R}\right) + w_{10}^{(4)}(z) + \mathcal{R}w_{10}^{(3)}(z) = \mathcal{R}\alpha^2 w_{10}'(z) + w_{10}''(z)\left(2\alpha^2 + i\omega_0\mathcal{R}\right), \quad \text{(A1)}$$

$$C_{l10}(z)\left(\alpha^2 + i\omega_0\right) = C_{l10}''(z) + C_{l10}'(z), \quad \text{(A2)}$$

and are subject to the boundary conditions

$$w_{10}(0) = 0, \quad w_{10}'(0) = i\eta\alpha_1\mathcal{R}, \quad (A3a,b)$$

$$w_{10}(z), w_{10}'(z), C_{l10}(z) \to 0 \quad \text{as} \quad z \to \infty, \quad (A4a-c)$$

$$\delta\eta\left(\beta + k_E + i\sigma_0\right) = (1+\beta)C_{l10}'(0) + \quad \text{(A5)}$$

$$+(1-k_E)C_{l10}(0), \tag{A6}$$

$$i\beta\delta\eta\sigma_0\mathcal{L} = \left(\mathcal{L}\left(\beta+k_E\right)+1-k_E\right)\left(C_{l10}(0)+\delta\eta\right)+ \\ +\left(\beta+1\right)\eta\left(k_E-1\right)\left(\alpha^2\Gamma+m_0+i\sigma_0\mathcal{U}\right), \tag{A7}$$

where

$$\mathcal{L} = \log\left[(\beta+k_E)/(\beta k_E+k_E)\right]. \tag{A8}$$

This zeroth-order problem (in $\mathcal{V}$) has been examined by [25]. An explicit solution for m_0 exists and is given by (45).

To proceed with higher order terms, it will be of use to note the form of the zeroth-order perturbed vertical velocity and solutal concentration fields, which are given by

$$w_{10}(z) = A_1 e^{-(\mathcal{R}+\lambda_2)z/2} + A_2 e^{-\alpha z}, \tag{A9}$$

$$C_{l10} = A_3 e^{-(\lambda_1+1)z/2}, \tag{A10}$$

where

$$A_1 = -A_2 = \frac{-2i\eta\alpha_1\mathcal{R}}{-2\alpha+\lambda_2+\mathcal{R}}, \tag{A11}$$

$$A_3 = -\frac{2\delta\eta\left(\beta+k_E+i\sigma_0\right)}{(\beta+1)\lambda_1+\beta+2k_E-1}, \tag{A12}$$

$$\lambda_2 = \sqrt{4\alpha^2+\mathcal{R}^2+4i\omega_0\mathcal{R}}. \tag{A13}$$

2. First order

The first-order perturbed vertical velocity w_{11} and solutal field C_{l11} satisfy the forced equations

$$\begin{aligned}
&-\alpha^2\left(\alpha^2+i\sigma_0\mathcal{R}\right)w_{11}(z)+\alpha^2\mathcal{R}w_{11}'(z) \\
&+\left(2\alpha^2+i\sigma_0\mathcal{R}\right)w_{11}''(z)-\mathcal{R}w_{11}{}^{(3)}(z)-w_{11}{}^{(4)}(z) \\
&=-i\mathcal{R}\left(k_1e^{-z\mathcal{R}}\left(\alpha^2+\mathcal{R}^2\right)-\alpha^2\left(k_1+\sigma_1\right)\right)w_{10}(z) \\
&-i\mathcal{R}\left(\sigma_1+k_1-k_1e^{-z\mathcal{R}}\right)w_{10}''(z)
\end{aligned} \tag{A14}$$

and

$$-\left(\alpha^2+i\sigma_0\right)C_{l11}(z)+C_{l11}'(z)+C_{l11}''(z)= \\ i\left(\sigma_1+k_1-k_1e^{-z\mathcal{R}}\right)C_{l10}(z)+\delta e^{-z}w_{10}(z), \tag{A15}$$

along with the boundary conditions

$$w_{11}(0)=0, \quad w_{11}'(0)=0, \tag{A16a,b}$$

$$w_{11}(z), w_{11}'(z), C_{l11}(z), \to 0 \quad \text{as} \quad z\to\infty, \tag{A17$a-c$}$$

$$(1+\beta)C_{l11}'(0)+(1-k_E)\,C_{l11}(0)=i\eta\sigma_1\delta \tag{A18}$$

where m_1 is determined by

$$i\beta\delta\eta\sigma_1\mathcal{L} = C_{l11}(0)\left(\mathcal{L}\left(\beta+k_E\right)-k_E+1\right) \\ +\left(\beta+1\right)\eta\left(k_E-1\right)\left(m_1+i\sigma_1\mathcal{U}\right), \tag{A19}$$

and ω_1 is chosen to make $\Im(m_1)=0$.

The solutions w_{11} and C_{l11} are of the form

$$w_{11}(z) = L_{11}e^{-\alpha z}+L_{12}e^{-z(3\mathcal{R}+\lambda_2)/2} \\ +(L_{13}+L_{23}z)e^{-z(\mathcal{R}+\lambda_2)/2}+L_{14}e^{-z(\alpha+\mathcal{R})}, \tag{A20}$$

$$\begin{aligned}
C_{l11}(z) = &\,P_{11}e^{-z(\alpha+1)}+P_{12}e^{-z(\lambda_2+\mathcal{R}+2)/2} \\
&+P_{13}e^{-z(\lambda_1+2\mathcal{R}+1)/2} \\
&+(P_{14}+P_{24}z)e^{-(\lambda_1+1)z/2}.
\end{aligned} \tag{A21}$$

The constants are given by

$$L_{12} = 2^{-1}A_1\alpha_1\left(-i\lambda_2+2\sigma_0+i\mathcal{R}\right)(6\alpha^2 \\ +4\mathcal{R}^2+\lambda_2\left(4\mathcal{R}+i\sigma_0\right)+7i\sigma_0\mathcal{R})^{-1}, \tag{A22}$$

$$L_{23} = (-iA_1\mathcal{R}^2\left(\alpha_1+\sigma_1\right)\left(\lambda_2+2i\sigma_0+\mathcal{R}\right))(\mathcal{R}\cdot \\ (4\alpha^2+\mathcal{R}^2+4i\sigma_0\mathcal{R})+\lambda_2\mathcal{R}(\mathcal{R}+2i\sigma_0))^{-1}, \tag{A23}$$

$$P_{12} = 2A_1\delta(\lambda_2(\mathcal{R}+1)+2i\sigma_0(\mathcal{R}-1) \\ +\mathcal{R}(\mathcal{R}+1))^{-1}, \tag{A24}$$

$$L_{14} = iA_2\alpha_1\mathcal{R}((\alpha-i\sigma_0)\left(2\alpha+\mathcal{R}\right))^{-1}, \tag{A25}$$

$$P_{11} = A_2\delta(\alpha-i\sigma_0)^{-1}, \tag{A26}$$

$$P_{13} = -iA_3\alpha_1(\mathcal{R}(\lambda_1+\mathcal{R}))^{-1}, \tag{A27}$$

$$P_{24} = -iA_3\left(\alpha_1+\sigma_1\right)\lambda_1^{-1}, \tag{A28}$$

$$L_{11} = (2L_{12}\mathcal{R}+2L_{14}\mathcal{R}-2L_{23})(-2\alpha+\lambda_2 \tag{A29}$$

$$+\mathcal{R})^{-1}-L_{14}, \tag{A30}$$

$$L_{13} = (-2L_{12}\mathcal{R}-2L_{14}\mathcal{R}+2L_{23})(-2\alpha+\lambda_2 \\ +\mathcal{R})^{-1}-L_{12}, \tag{A31}$$

$$\begin{aligned}
P_{14} = &-P_{13}+\Big[-2i\delta\eta\sigma_1+2(\beta+1)P_{24} \\
&-2P_{11}\gamma_1+P_{12}\gamma_2-2(\beta+1)P_{13}\mathcal{R}\Big]\cdot \\
&\cdot\Big[(\beta+1)\lambda_1+\beta+2k_E-1\Big]^{-1},
\end{aligned} \tag{A32}$$

where

$$\gamma_1 = \alpha(\beta+1)+(\beta+k_E), \tag{A33}$$

$$\gamma_2 = -(\beta+1)(\lambda_2+\mathcal{R})-2(\beta+k_E). \tag{A34}$$

Noting that $C_{l11}(z)$ is pure imaginary if $\omega_0=0$, we deduce from the real part of (A19) that $m_1=0$. If $\omega_0\neq 0$, then we deduce from (A19) that m_1 is given by

$$m_1 = \frac{\mathcal{L}\left(\beta+k_E\right)+\left(1-k_E\right)}{\eta(\beta+1)\left(1-k_E\right)}\sum_{j=1}^{4}\Re(P_{1j}). \tag{A35}$$

3. Second order

It is necessary to examine second-order asymptotics to determine the influence of flow on the leading-order steady branch.

The governing equations for the perturbed vertical velocity and solutal fields, w_{12} and C_{l12}, are given by the forced differential equations

$$\begin{aligned} &{w_{12}}^{(4)}(z) + \mathcal{R}{w_{12}}^{(3)}(z) - \left(2\alpha^2 + i\sigma_0\mathcal{R}\right) w_{12}''(z) \\ &-\alpha^2\mathcal{R}w_{12}'(z) + \left(\alpha^4 + i\alpha^2\sigma_0\mathcal{R}\right) w_{12}(z) = \\ &\qquad = H_{112}e^{-z(\alpha+2\mathcal{R})} + H_{113}e^{-z(\alpha+\mathcal{R})} \\ &\qquad + H_{111}e^{-z(\lambda_2+5\mathcal{R})/2} \\ &\qquad + (H_{114} + zH_{124})e^{-z(\lambda_2+3\mathcal{R})/2} \\ &\qquad + (H_{115} + zH_{125})e^{-z(\lambda_2+\mathcal{R})/2}, \end{aligned} \tag{A36}$$

$$\begin{aligned} &C_{l12}''(z) + C_{l12}'(z) - (\alpha^2 + i\sigma_0)C_{l12}(z) = \\ &\quad = H_{211}e^{-z(\alpha+\mathcal{R}+1)} + H_{212}e^{-(\alpha+1)z} \\ &\quad + H_{213}e^{-z(\lambda_2+3\mathcal{R}+2)/2} \\ &\quad + (zH_{227} + H_{217})e^{-(\lambda_1+1)z/2} \\ &\quad + (zH_{224} + H_{214})e^{-z(\lambda_2+\mathcal{R}+2)/2} \\ &\quad + H_{215}e^{-z(\lambda_1+4\mathcal{R}+1)/2} \\ &\quad + (zH_{226} + H_{216})e^{-z(\lambda_1+2\mathcal{R}+1)/2}. \end{aligned} \tag{A37}$$

The forcing terms involve lower-order solutions and can be succintly given in terms of the following constants

$$\begin{aligned} H_{111} =&\frac{1}{2}\alpha_1\mathcal{R}^2L_{12}(-3i\lambda_2 + 2\sigma_0 - 3i\mathcal{R}), \\ H_{112} =&-2i\alpha\alpha_1\mathcal{R}^2L_{14}, \\ H_{113} =&i\mathcal{R}^2(L_{14}(\alpha_1+\sigma_1)(2\alpha+\mathcal{R}) + \alpha_1\mathcal{R}L_{11}), \\ H_{114} =&\frac{1}{2}i\mathcal{R}\Big(\lambda_2(3\mathcal{R}(\alpha_1+\sigma_1)L_{12} - \alpha_1\mathcal{R}L_{13} \\ &+ 2\alpha_1L_{23}) + \mathcal{R}((\alpha_1+\sigma_1)(5\mathcal{R}+2i\sigma_0)L_{12} \\ &+ \alpha_1(\mathcal{R}-2i\sigma_0)L_{13} + 2\alpha_1L_{23})\Big), \\ H_{115} =&\frac{1}{2}i\mathcal{R}^2\left(\lambda_2 + 2i\sigma_0 + \mathcal{R}\right)\left(L_{13}\left(\alpha_1+\sigma_1\right)\right. \\ &\left.+ A_1\sigma_2\right) - i\mathcal{R}L_{23}\left(\alpha_1+\sigma_1\right)\left(\lambda_2+\mathcal{R}\right), \\ H_{124} =&\frac{1}{2}\alpha_1\mathcal{R}^2L_{23}(-i\lambda_2 + 2\sigma_0 + i\mathcal{R}), \\ H_{125} =&\frac{1}{2}i\mathcal{R}^2L_{23}(\alpha_1+\sigma_1)(\lambda_2 + 2i\sigma_0 + \mathcal{R}), \\ H_{211} =&\delta L_{14} - i\alpha_1P_{11}, \\ H_{212} =&\delta L_{11} + iP_{11}(\alpha_1+\sigma_1), \\ H_{213} =&\delta L_{12} - i\alpha_1P_{12}, \\ H_{214} =&\delta L_{13} + iP_{12}(\alpha_1+\sigma_1), \\ H_{215} =&-i\alpha_1P_{13}, \quad H_{224} = \delta L_{23}, \\ H_{216} =&i(P_{13}(\alpha_1+\sigma_1) - \alpha_1P_{14}), \\ H_{217} =&i(P_{14}(\alpha_1+\sigma_1) + A_3\sigma_2), \\ H_{226} =&-i\alpha_1P_{24}, \quad H_{227} = iP_{24}(\alpha_1+\sigma_1). \end{aligned} \tag{A38}$$

The corresponding boundary conditions are

$$w_{12}(0) = 0, \quad w_{12}'(0) = 0, \tag{$A39a,b$}$$

$$w_{12}(z), w_{12}'(z), c_{l12}(z) \to 0, \quad \text{as} \quad z \to \infty, \tag{$A40a-c$}$$

$$(1-k_E)\,c_{l12}(0) + (\beta+1)c_{l12}'(0) - i\delta\eta\sigma_2 = 0. \tag{A41}$$

The solutions are given by

$$\begin{aligned} w_{12}(z) =& J_{111}e^{-z(\alpha+\mathcal{R})} + J_{112}e^{-z(\alpha+2\mathcal{R})} \\ &+ J_{113}e^{-z(\lambda_2+5\mathcal{R})/2} + J_{116}e^{-\alpha z} \\ &+ (J_{114} + zJ_{124})e^{-z(\lambda_2+3\mathcal{R})/2} \\ &+ \left(J_{115} + zJ_{125} + z^2J_{135}\right)e^{-z(\lambda_2+\mathcal{R})/2}, \end{aligned} \tag{A42}$$

$$\begin{aligned} C_{12}(z) =& J_{211}e^{-(\alpha+1)z} + J_{212}e^{-z(\alpha+\mathcal{R}+1)} \\ &+ J_{213}e^{-z(\lambda_1+4\mathcal{R}+1)/2} \\ &+ J_{214}e^{-z(\lambda_2+3\mathcal{R}+2)/2} \\ &+ (zJ_{225} + J_{215})e^{-\frac{1}{2}z(\lambda_2+\mathcal{R}+2)} \\ &+ (zJ_{226} + J_{216})e^{-\frac{1}{2}z(\lambda_1+2\mathcal{R}+1)} \\ &+ \left(J_{217} + zJ_{227} + z^2J_{237}\right)e^{-(\lambda_1+1)z/2}. \end{aligned} \tag{A43}$$

We note that both of these satisfy the required decay conditions despite the polynomial terms, owing to the sign of the exponential decay constants. The remaining constants are given by

$$\begin{aligned} J_{111} =&H_{113}(\mathcal{R}^2\alpha(2\alpha+\mathcal{R}))^{-1}, \\ J_{112} =&H_{112}(4\mathcal{R}^2(\alpha+\mathcal{R})\left(3\alpha+2\mathcal{R}\right))^{-1}, \\ J_{113} =&H_{111}(\mathcal{R}^2\left(20\alpha^2 + 31\mathcal{R}^2 + 23\mathcal{R}\lambda_2\right))^{-1}, \\ J_{124} =&H_{124}(\mathcal{R}^2\left(6\alpha^2 + 4\mathcal{R}^2 + 4\mathcal{R}\lambda_2\right))^{-1}, \\ J_{114} =&\Big[H_{114}\left(2\mathcal{R}\left(6\alpha^2 + 4\mathcal{R}^2\right) + 8\lambda_2\mathcal{R}^2\right) \\ &+ H_{124}\left(20\alpha^2 + 19\mathcal{R}\lambda_2 + 21\mathcal{R}^2\right)\Big]\cdot \\ &\cdot\left[2\mathcal{R}^3\left(6\alpha^2 + 4\mathcal{R}^2 + 4\mathcal{R}\lambda_2\right)^2\right]^{-1}, \\ J_{125} =&\Big[\left(-2\mathcal{R}\left(4\alpha^2 + \mathcal{R}^2\right) - 2\mathcal{R}^2\lambda_2\right)H_{115} \\ &+ \left(-6\lambda_2\mathcal{R} - 2\left(8\alpha^2 + 3\mathcal{R}^2\right)\right)H_{125}\Big]\cdot \\ &\cdot\left[\mathcal{R}^2\left(4\alpha^2 + \mathcal{R}^2 + \mathcal{R}\lambda_2\right)^2\right]^{-1}, \\ J_{135} =&-H_{125}(\mathcal{R}\left(4\alpha^2 + \mathcal{R}^2\right) + \mathcal{R}^2\lambda_2)^{-1}, \\ J_{211} =&H_{212}\alpha^{-1}, \\ J_{212} =&H_{211}(\alpha + \mathcal{R}^2 + 2\alpha\mathcal{R} + \mathcal{R})^{-1}, \\ J_{213} =&H_{215}(2\mathcal{R}\left(\lambda_1 + 2\mathcal{R}\right))^{-1}, \\ J_{214} =&2H_{213}(\lambda_2(3\mathcal{R}+1) + \mathcal{R}(5\mathcal{R}+3))^{-1}, \\ J_{215} =&4H_{224}(\lambda_2 + (\mathcal{R}+1))(\lambda_2+\mathcal{R})^{-2}(\mathcal{R}+1)^{-2} \\ &+ 2H_{214}(\lambda_2+\mathcal{R})^{-1}(\mathcal{R}+1)^{-1}, \\ J_{225} =&2H_{224}(\lambda_2+\mathcal{R})^{-1}(\mathcal{R}+1)^{-1}, \\ J_{237} =&-2^{-1}H_{227}\lambda_1^{-1} \end{aligned}$$

$$
\begin{aligned}
J_{216} =& H_{216}(\mathcal{R}\,(\lambda_1+\mathcal{R}))^{-1} \\
&+ H_{226}\,(\lambda_1+2\mathcal{R})\,(\mathcal{R}^2\,(\lambda_1+\mathcal{R})^2)^{-1}, \\
J_{226} =& H_{226}(\mathcal{R}\,(\lambda_1+\mathcal{R}))^{-1}, \\
J_{227} =& -H_{217}\lambda_1^{-1} - H_{227}\lambda_1^{-2},
\end{aligned} \tag{A44}
$$

and

$$
\begin{aligned}
J_{115} =& \Big[-2\mathcal{R}J_{111} - 4\mathcal{R}J_{112} + 2J_{124} + 2J_{125} \\
& (2\alpha - 5\mathcal{R} - \lambda_2)J_{113} + (2\alpha - 3\mathcal{R} - \lambda_2)J_{114}\Big] \cdot \\
& \cdot (-2\alpha + \lambda_2 + \mathcal{R})^{-1}, \\
J_{116} =& \Big[(2\alpha + \mathcal{R} - \lambda_2)J_{111} + (2\alpha + 3\mathcal{R} - \lambda_2)J_{112} \\
& + 4\mathcal{R}J_{113} + 2\mathcal{R}J_{114} - 2J_{124} - 2J_{125}\Big] \cdot \\
& \cdot (-2\alpha + \lambda_2 + \mathcal{R})^{-1}, \\
J_{217} =& \Big[(1-k_E)\,(J_{213}+J_{216}) - (\beta+k_E)\,(2J_{211} \\
& + 2J_{212} + J_{213} + 2J_{214} + 2J_{215} + J_{216}) \\
& + (\beta+1)\Big(-2\alpha J_{211} - 2(\alpha+\mathcal{R})J_{212} \\
& - (\lambda_1 + 4\mathcal{R})\,J_{213} - (\lambda_2 + 3\mathcal{R})\,J_{214} \\
& - (\lambda_2 + \mathcal{R})J_{215} - (\lambda_1 + 2\mathcal{R})\,J_{216} + 2J_{225} \\
& + 2J_{226} + 2J_{227}\Big) - 2i\delta\eta\sigma_2\Big] \cdot \\
& \cdot ((\beta+1)\lambda_1 + \beta + 2k_E - 1)^{-1},
\end{aligned} \tag{A45}
$$

For brevity, we have set $\omega_0 = 0$, corresponding to the leading-order steady branch.

The second-order correction m_2 is determined by

$$
\begin{aligned}
i\beta\delta\eta\sigma_2\mathcal{L} \;=& \; C_{l12}(0)\,(\mathcal{L}\,(\beta+k_E) + 1 - k_E) \\
& - \eta(\beta+1)\,(1-k_E)\,(m_2 + i\sigma_2\mathcal{U})\,,
\end{aligned} \tag{A46}
$$

once the solutal field C_{l12} is known and the value of σ_2 is determined by requiring that $\Im(m_2) = 0$. This gives

$$
m_2 = \frac{\mathcal{L}\,(\beta+k_E) + (1-k_E)}{\eta(\beta+1)\,(1-k_E)} \sum_{j=1}^{7} \Re(J_{21j}), \tag{A47}
$$

thus determining the influence of weak flow on the steady branch.

Appendix B: Singular perturbations

1. Zeroth order in $\mathcal{V}^{1/3}$

The governing equations for the perturbed vertical velocity and solutal fields correspond to those of the zeroth-order regular expansion in Sec. A 1, in which it is taken that $\omega_0 = 0$ and $\alpha = \alpha_1 = \alpha_{10}$. The solution for the two fields is given by (A10), where the constants $A_1, A_2, A_3, \lambda_1, \lambda_2$ are modified in that $\omega_0 = 0$ and $\alpha = \alpha_1 = \alpha_{10}$. The zeroth-order contribution to the inverse morphological number may be simplified to

$$
\begin{aligned}
\tilde{m}_0 =& -\alpha_{10}^2\Gamma + \frac{\delta(1-k_E) + \mathcal{L}\,(\beta+k_E)}{(\beta+1)\,(1-k_E)} \cdot \\
& \cdot \left(1 - \frac{2\,(\beta+k_E)}{(\beta+1)\lambda_1 + \beta + 2k_E - 1}\right).
\end{aligned} \tag{B1}
$$

2. First order in $\mathcal{V}^{1/3}$

At first-order in $\mathcal{V}^{1/3}$, the governing equations reduce to

$$
\begin{aligned}
& \tilde{w}_{11}{}^{(4)}(z) + \mathcal{R}\tilde{w}_{11}{}^{(3)}(z) - 2\alpha_{10}^2\tilde{w}_{11}''(z) - \alpha_{10}^2\mathcal{R}\tilde{w}_{11}'(z) \\
& + \alpha_{10}^4\tilde{w}_{11}(z) = -i\alpha_{10}^2\mathcal{R}\tilde{\omega}_1\tilde{w}_{10}(z) + i\mathcal{R}\tilde{\omega}_1\tilde{w}_{10}''(z),
\end{aligned} \tag{B2}
$$

$$
\tilde{c}_{l11}''(z) + \tilde{c}_{l11}'(z) - \alpha_{10}^2\tilde{c}_{l11}(z) = i\tilde{\omega}_1\tilde{c}_{l10}(z), \tag{B3}
$$

and are subject to the boundary conditions

$$
\tilde{w}_{11}(0) = 0, \quad \tilde{w}_{11}'(0) = 0, \tag{$B4a,b$}
$$

$$
\tilde{w}_{11}(z), \tilde{w}_{11}'(z), \tilde{c}_{l11}(z) \to 0 \quad \text{as} \quad z \to \infty, \tag{$B5a-c$}
$$

$$
(\beta+1)\tilde{c}_{l11}'(0) + (1-k_E)\,\tilde{c}_{l11}(0) - i\delta\eta\tilde{\omega}_1 = 0, \tag{B6}
$$

$$
i\beta\delta\eta\mathcal{L}\tilde{\omega}_1 \;= \tilde{c}_{l11}(0)\,(\mathcal{L}\,(\beta+k_E) + (1-k_E)) \tag{B7}
$$

$$
+ (\beta+1)\,(k_E-1)\,i\eta\mathcal{U}\tilde{\omega}_1. \tag{B8}
$$

This is a forced system of differential equations, where the forcing term is given in terms of the zeroth-order solutions. Its solution is given by

$$
\tilde{w}_{11} = \tilde{L}_{1,2}e^{-\alpha_{10}z} + \left(z\tilde{L}_{2,1} + \tilde{L}_{1,1}\right)e^{-\frac{1}{2}z\left(\tilde{\lambda}_2 + \mathcal{R}\right)}, \tag{B9}
$$

$$
\tilde{c}_{l11} = \left(z\tilde{P}_{2,1} + \tilde{P}_{1,1}\right)e^{-\frac{1}{2}z\left(\tilde{\lambda}_1 + 1\right)}, \tag{B10}
$$

where

$$
\tilde{L}_{1,1} = -\tilde{L}_{1,2} = -\frac{2i\mathcal{R}\tilde{A}_1\tilde{\omega}_1}{\left(\tilde{\lambda}_2 - 2\alpha_{10} + \mathcal{R}\right)\tilde{\lambda}_2}, \tag{B11}
$$

$$
\tilde{L}_{2,1} = -i\mathcal{R}\tilde{A}_1\tilde{\omega}_1/\tilde{\lambda}_2, \tag{B12}
$$

$$
\tilde{P}_{1,1} = -2i\tilde{\omega}_1\frac{(\beta+1)\tilde{A}_3/\tilde{\lambda}_1 + \delta\eta}{(\beta+1)\tilde{\lambda}_1 + \beta + 2k_E - 1}, \tag{B13}
$$

$$
\tilde{P}_{2,1} = -i\tilde{A}_3\tilde{\omega}_1/\tilde{\lambda}_1, \tag{B14}
$$

and

$$
\tilde{\lambda}_1 = \sqrt{4\alpha_{10}^2 + 1}, \quad \tilde{\lambda}_2 = \sqrt{4\alpha_{10}^2 + \mathcal{R}^2}. \tag{$B15a,b$}
$$

We find the first-order restriction on the inverse morphological number is given by

$$i\mathcal{U}\tilde{\omega}_1 = \frac{\tilde{P}_{1,1}\left(\mathcal{L}\left(\beta + k_E\right) + \left(1 - k_E\right)\right) - i\beta\delta\eta\mathcal{L}\tilde{\omega}_1}{\left(\beta + 1\right)\left(1 - k_E\right)\eta}. \tag{B16}$$

Noting that $\tilde{A}_3$ is real, and hence that $\tilde{P}_{1,1}$ is pure imaginary, it must be that all of the terms above are purely imaginary. This ensures that the first-order contribution to the inverse morphological number is real, which we expect on physical grounds. This relation gives that either $\tilde{\omega}_1 = 0$, which we exclude as it corresponds to the regular expansion away from the singular root, or that the wavenumber α_{10} satisfies the relation (63).

Higher orders need to be considered to restrict $\tilde{\omega}_1$. We will find that examining the third-order in $\mathcal{V}^{1/3}$ is sufficient.

3. Second order in $\mathcal{V}^{1/3}$

The governing equations at $\mathcal{O}(\mathcal{V}^{2/3})$ are

$$\begin{aligned}\tilde{w}_{12}{}^{(4)}(z) &+ \mathcal{R}\tilde{w}_{12}{}^{(3)}(z) - 2\alpha_{10}^2\tilde{w}_{12}''(z) - \alpha_{10}^2\mathcal{R}\tilde{w}_{12}'(z) \\ &+\alpha_{10}^4\tilde{w}_{12}(z) = -\alpha_{10}^2\tilde{w}_{10}(z)\left(4\alpha_{10}\alpha_{11} + i\mathcal{R}\tilde{\omega}_2\right) \\ &\quad + 2\alpha_{11}\alpha_{10}\mathcal{R}\tilde{w}_{10}'(z) + 4\alpha_{11}\alpha_{10}\tilde{w}_{10}''(z) \\ &\quad + i\mathcal{R}\tilde{\omega}_2\tilde{w}_{10}''(z) - i\alpha_{10}^2\mathcal{R}\tilde{\omega}_1\tilde{w}_{11}(z) \\ &\quad + i\mathcal{R}\tilde{\omega}_1\tilde{w}_{11}''(z),\end{aligned} \tag{B17}$$

for the vertical velocity, and

$$\begin{aligned}\tilde{c}_{l12}''(z) + \tilde{c}_{l12}'(z) - \alpha_{10}^2\tilde{c}_{l12}(z) &= i\tilde{\omega}_1\tilde{c}_{l11}(z) \\ &\quad + \left(2\alpha_{10}\alpha_{11} + i\tilde{\omega}_2\right)\tilde{c}_{l10}(z),\end{aligned} \tag{B18}$$

for the solutal field. The boundary conditions reduce to

$$\tilde{w}_{12}(0) = 0, \quad \tilde{w}_{12}'(0) = i\eta\alpha_{11}\mathcal{R}, \tag{B19a, b}$$

$$\tilde{w}_{12}(z), \tilde{w}_{12}'(z), \tilde{c}_{l12}(z) \to 0, \quad \text{as} \quad z \to \infty, \tag{B20a-c}$$

$$(\beta + 1)\tilde{c}_{l12}'(0) + \left(1 - k_E\right)\tilde{c}_{l12}(0) = i\delta\eta\tilde{\omega}_2, \tag{B21}$$

$$\begin{aligned}&\tilde{c}_{l12}(0)\left(\mathcal{L}(k_E + \beta) + 1 - k_E\right)/\eta = i\beta\delta\mathcal{L}\tilde{\omega}_2 \\ &\quad - \left(\beta + 1\right)\left(k_E - 1\right)\left(\tilde{m}_2 + i\mathcal{U}\tilde{\omega}_2 + 2\Gamma\alpha_{10}\alpha_{11}\right).\end{aligned} \tag{B22}$$

The solution is given by

$$\begin{aligned}\tilde{w}_{12}(z) &= \left(z^2\tilde{J}_{1,3,1} + z\tilde{J}_{1,2,1} + \tilde{J}_{1,1,1}\right)e^{-z\left(\tilde{\lambda}_2 + \mathcal{R}\right)/2} \\ &\quad + \left(z\tilde{J}_{1,2,2} + \tilde{J}_{1,1,2}\right)e^{-\alpha_{10}z}, \\ \tilde{c}_{l12}(z) &= \left(z^2\tilde{J}_{2,3,1} + z\tilde{J}_{2,2,1} + \tilde{J}_{2,1,1}\right)e^{-z\left(\tilde{\lambda}_1 + 1\right)/2},\end{aligned} \tag{B23}$$

where

$$\begin{aligned}\tilde{J}_{1,2,1} =& -2\left[\tilde{\lambda}_2\mathcal{R}\tilde{H}_{1,1,1} + (2\tilde{\lambda}_2 + \mathcal{R})\tilde{H}_{1,2,1}\right]\cdot \\ &\cdot\left[\tilde{\lambda}_2^2\mathcal{R}^2(\tilde{\lambda}_2 + \mathcal{R})\right]^{-1}, \\ \tilde{J}_{1,2,2} =& 2^{-1}\tilde{H}_{1,1,2}\alpha_{10}^{-2}\mathcal{R}^{-1}, \\ \tilde{J}_{2,2,1} =& -\tilde{H}_{2,1,1}\tilde{\lambda}_1^{-1} - \tilde{H}_{2,2,1}\tilde{\lambda}_1^{-2}, \\ \tilde{J}_{2,3,1} =& -2^{-1}\tilde{H}_{2,2,1}\tilde{\lambda}_1^{-1}, \\ \tilde{J}_{1,3,1} =& -\tilde{H}_{1,2,1}(\mathcal{R}\tilde{\lambda}_2(\mathcal{R} + \tilde{\lambda}_2))^{-1}, \\ \tilde{J}_{1,1,1} =& -\tilde{J}_{1,1,2} = 2(\tilde{J}_{1,2,1} + \tilde{J}_{1,2,2} - i\eta\alpha_{11}\mathcal{R})\cdot \\ &\cdot(\tilde{\lambda}_2 - 2\alpha_{10} + \mathcal{R})^{-1}, \\ \tilde{J}_{2,1,1} =& 2\left[(\beta + 1)\tilde{J}_{2,2,1} - i\delta\eta\tilde{\omega}_2\right]\cdot \\ &\cdot\left[(\beta + 1)\tilde{\lambda}_1 + \beta + 2k_E - 1\right]^{-1},\end{aligned} \tag{B24}$$

and

$$\begin{aligned}\tilde{H}_{1,1,1} =& \mathcal{R}(\tilde{\lambda}_2 + \mathcal{R})\left[2\alpha_{10}\alpha_{11}\tilde{A}_1 + i(\mathcal{R}\tilde{\omega}_1\tilde{L}_{1,1}\right. \\ &\left. + \mathcal{R}\tilde{A}_1\tilde{\omega}_2 - 2\tilde{\omega}_1\tilde{L}_{2,1})\right]/2, \\ \tilde{H}_{1,2,1} =& i\mathcal{R}^2\tilde{\omega}_1\tilde{L}_{2,1}(\tilde{\lambda}_2 + \mathcal{R})/2, \\ \tilde{H}_{1,1,2} =& -2\alpha_{10}\mathcal{R}(\alpha_{10}\alpha_{11}\tilde{A}_2 + i\tilde{\omega}_1\tilde{L}_{2,2}), \\ \tilde{H}_{2,1,1} =& 2\alpha_{10}\alpha_{11}\tilde{A}_3 + i(\tilde{\omega}_1\tilde{P}_{1,1} + \tilde{A}_3\tilde{\omega}_2), \\ \tilde{H}_{2,2,1} =& i\tilde{\omega}_1\tilde{P}_{2,1},\end{aligned} \tag{B25}$$

from which we deduce that

$$\begin{aligned}\tilde{m}_2 = -2\Gamma\alpha_{10}\alpha_{11} - i\mathcal{U}\tilde{\omega}_2 - &\left[\tilde{J}_{2,1,1}(\mathcal{L}(k_E + \beta) + 1\right. \\ &\left. - k_E) - i\beta\delta\eta\mathcal{L}\tilde{\omega}_2\right]\left[(\beta + 1)\left(k_E - 1\right)\eta\right]^{-1},\end{aligned} \tag{B26}$$

Requiring $\tilde{m}_2$ to be real, we find that $\tilde{m}_2$ satisfies (67) and $\tilde{\omega}_2$ solves

$$\begin{aligned}&\beta\delta\mathcal{L}\eta\tilde{\omega}_2 - \mathcal{U}(\beta + 1)\left(k_E - 1\right)\eta\tilde{\omega}_2 = \\ &\qquad \Im(\tilde{J}_{2,1,1})\left(\mathcal{L}(k_E + \beta) + 1 - k_E\right).\end{aligned} \tag{B27}$$

Since $\tilde{m}_2$ depends on $\tilde{\omega}_1$ through $\Re\left(\tilde{J}_{2,1,1}\right)$, we note that we have insufficient information to compute the value of $\tilde{m}_2$, as no information about $\tilde{\omega}_1$ is revealed up to the current order in $\mathcal{V}^{1/3}$. It is necessary to examine the next higher order for this.

4. Third order in $\mathcal{V}^{1/3}$

The governing equations at third-order in $\mathcal{V}^{1/3}$ are

$$\begin{aligned}&\tilde{w}_{13}{}^{(4)}(z) + \mathcal{R}\tilde{w}_{13}{}^{(3)}(z) - 2\alpha_{10}^2\tilde{w}_{13}''(z) - \alpha_{10}^2\mathcal{R}\tilde{w}_{13}'(z) \\ &\quad + \alpha_{10}^4\tilde{w}_{13}(z) = i\mathcal{R}\tilde{\omega}_1\tilde{w}_{12}''(z) - i\alpha_{10}^2\mathcal{R}\tilde{\omega}_1\tilde{w}_{12}(z) \\ &\quad + \left(4\alpha_{10}\alpha_{11} + i\mathcal{R}\tilde{\omega}_2\right)\tilde{w}_{11}''(z) + 2\alpha_{11}\alpha_{10}\mathcal{R}\tilde{w}_{11}'(z)\end{aligned}$$

$$
\begin{aligned}
&-\alpha_{10}^2\tilde{w}_{11}(z)\left(4\alpha_{10}\alpha_{11}+i\mathcal{R}\tilde{\omega}_2\right)+i\mathcal{R}\tilde{w}_{10}''(z)\cdot\\
&\cdot\left(\alpha_{10}(1-e^{-z\mathcal{R}})+\tilde{\omega}_3\right)+i\Big((\alpha_{10}^2+\mathcal{R}^2)e^{-z\mathcal{R}}\\
&-\left(2\alpha_{11}\tilde{\omega}_1+\alpha_{10}\left(\alpha_{10}+\sigma 3\right)\right)\Big)\alpha_{10}\mathcal{R}\tilde{w}_{10}(z), \quad \text{(B28)}
\end{aligned}
$$

for the velocity field and

$$
\tilde{c}_{l13}''(z)+\tilde{c}_{l13}'(z)-\alpha_{10}^2\tilde{c}_{l13}(z)=\delta e^{-z}\tilde{w}_{10}(z) \quad \text{(B29)}
$$

$$
\begin{aligned}
&+i\tilde{c}_{l10}(z)\left(\tilde{\omega}_3+\alpha_{10}\left(1-e^{-z\mathcal{R}}\right)\right)\\
&+\left(2\alpha_{10}\alpha_{11}+i\tilde{\omega}_2\right)\tilde{c}_{l11}(z)+i\tilde{\omega}_1\tilde{c}_{l12}(z), \quad \text{(B30)}
\end{aligned}
$$

for the solutal field. The boundary conditions reduce to

$$
\tilde{w}_{13}(0)=0, \quad \tilde{w}_{13}'(0)=0,
$$

$$
\tilde{w}_{13}(z), \tilde{w}_{13}'(z), \tilde{c}_{l13}(z)\to 0 \quad \text{as} \quad z\to\infty
$$

$$
\left(1-k_E\right)\tilde{c}_{l13}(0)+(\beta+1)\tilde{c}_{l13}'(0)-i\delta\eta\tilde{\omega}_3=0, \quad \text{(B31)}
$$

$$
\begin{aligned}
\tilde{c}_{l13}(0)\left(\mathcal{L}(k_E+\beta)+1-k_E\right)/\eta=i\beta\delta\mathcal{L}\tilde{\omega}_3&\\
+(\beta+1)\left(1-k_E\right)\left(\tilde{m}_3+i\mathcal{U}\tilde{\omega}_3\right).& \quad \text{(B32)}
\end{aligned}
$$

The solution is forced by the solutions corresponding to the three lower orders and is given by

$$
\begin{aligned}
w_{13}(z) \;=\; &\tilde{\chi}_{1,1,1}e^{-\frac{1}{2}z\left(\tilde{\lambda}_2+3\mathcal{R}\right)}+\tilde{\chi}_{1,1,2}e^{-z(\alpha_{10}+\mathcal{R})}\\
&+\left(z^3\tilde{\chi}_{1,4,3}+z^2\tilde{\chi}_{1,3,3}+z\tilde{\chi}_{1,2,3}+\tilde{\chi}_{1,1,3}\right)e^{-\frac{1}{2}z\left(\tilde{\lambda}_2+\mathcal{R}\right)}\\
&+\left(z^2\tilde{\chi}_{1,3,4}+z\tilde{\chi}_{1,2,4}+\tilde{\chi}_{1,1,4}\right)e^{-\alpha_{10}z}, \quad \text{(B33)}\\
c_{l13}(z) \;=\; &\tilde{\chi}_{2,1,1}e^{-\frac{1}{2}z\left(\tilde{\lambda}_1+2\mathcal{R}+1\right)}\\
&+\left(z^3\tilde{\chi}_{2,4,4}+z^2\tilde{\chi}_{2,3,4}+z\tilde{\chi}_{2,2,4}+\tilde{\chi}_{2,1,4}\right)e^{-\frac{1}{2}z\left(\tilde{\lambda}_1+1\right)}\\
&+\tilde{\chi}_{2,1,2}e^{-\frac{1}{2}z\left(\tilde{\lambda}_2+\mathcal{R}+2\right)}+\tilde{\chi}_{2,1,3}e^{-(\alpha_{10}+1)z}, \quad \text{(B34)}
\end{aligned}
$$

where,

$$
\begin{aligned}
\tilde{\chi}_{1,1,1} =&\tilde{R}_{1,1,1}(2\mathcal{R}^2(2\mathcal{R}\tilde{\lambda}_2+3\alpha_{10}^2+2\mathcal{R}^2))^{-1},\\
\tilde{\chi}_{1,1,2} =&\tilde{R}_{1,1,2}(\alpha_{10}\mathcal{R}^2\left(2\alpha_{10}+\mathcal{R}\right))^{-1},\\
\tilde{\chi}_{1,2,3} =& 2\tilde{R}_{1,1,3}((\tilde{\lambda}_2+\mathcal{R})\mathcal{R}\tilde{\lambda}_2)^{-1}-2\tilde{R}_{1,2,3}(2\tilde{\lambda}_2+\mathcal{R})\cdot\\
&\cdot((\tilde{\lambda}_2+\mathcal{R})\mathcal{R}^2\tilde{\lambda}_2^2)^{-1}-4\tilde{R}_{1,3,3}(2\tilde{\lambda}_2+\mathcal{R})\cdot\\
&\cdot(2\tilde{\lambda}_2^2+\tilde{\lambda}_2\mathcal{R}+\mathcal{R}^2)(\mathcal{R}^3\tilde{\lambda}_2^3(\tilde{\lambda}_2+\mathcal{R})^2)^{-1},\\
\tilde{\chi}_{1,3,3} =& -2\tilde{R}_{1,3,3}(\mathcal{R}+2\tilde{\lambda}_2)(\mathcal{R}^2\tilde{\lambda}_2^2(\mathcal{R}+\tilde{\lambda}_2))^{-1}\\
&-\tilde{R}_{1,2,3}(\mathcal{R}\tilde{\lambda}_2(\mathcal{R}+\tilde{\lambda}_2))^{-1},\\
\tilde{\chi}_{1,4,3} =& -2\tilde{R}_{1,3,3}(3\mathcal{R}\tilde{\lambda}_2(\mathcal{R}+\tilde{\lambda}_2))^{-1},\\
\tilde{\chi}_{1,2,4} =&(2\alpha_{10}\mathcal{R}\tilde{R}_{1,1,4}+(3\mathcal{R}-4\alpha_{10})\tilde{R}_{1,2,4})\cdot\\
&\cdot(4\alpha_{10}^3\mathcal{R}^2)^{-1},\\
\tilde{\chi}_{1,3,4} =&\tilde{R}_{1,2,4}(4\alpha_{10}^2\mathcal{R})^{-1}, \quad \tilde{\chi}_{2,1,3}=\tilde{R}_{2,1,2}\alpha_{10}^{-1},\\
\tilde{\chi}_{2,1,1} =&\tilde{R}_{2,1,3}(\mathcal{R}(\tilde{\lambda}_1+\mathcal{R}))^{-1},\\
\tilde{\chi}_{2,1,2} =&2\tilde{R}_{2,1,1}((\mathcal{R}+1)(\tilde{\lambda}_2+\mathcal{R}))^{-1},\\
\tilde{\chi}_{2,2,4} =& -(4\alpha_{10}^2\tilde{R}_{2,1,4}+\tilde{\lambda}_1\tilde{R}_{2,2,4}+\tilde{R}_{2,1,4}\\
&+2\tilde{R}_{2,3,4})\tilde{\lambda}_1^{-3/2},\\
\tilde{\chi}_{2,3,4} =& -2^{-1}\tilde{R}_{2,2,4}\tilde{\lambda}_1^{-1}-\tilde{R}_{2,3,4}\tilde{\lambda}_1^{-2},\\
\tilde{\chi}_{2,4,4} =& -3^{-1}\tilde{R}_{2,3,4}\tilde{\lambda}_1^{-2}, \quad \text{(B35)}
\end{aligned}
$$

and

$$
\begin{aligned}
\tilde{\chi}_{1,1,3} =&(2\tilde{\chi}_{1,2,3}+2\tilde{\chi}_{1,2,4}-2\mathcal{R}\tilde{\chi}_{1,1,1}-2\mathcal{R}\tilde{\chi}_{1,1,2})\cdot\\
&\cdot(\tilde{\lambda}_2-2\alpha_{10}+\mathcal{R})^{-1}-\tilde{\chi}_{1,1,1},\\
\tilde{\chi}_{1,1,4} =&(2\mathcal{R}\tilde{\chi}_{1,1,1}+2\mathcal{R}\tilde{\chi}_{1,1,2}-2\tilde{\chi}_{1,2,3}-2\tilde{\chi}_{1,2,4})\cdot\\
&\cdot(\tilde{\lambda}_2-2\alpha_{10}+\mathcal{R})^{-1}-\tilde{\chi}_{1,1,2},\\
\tilde{\chi}_{2,1,4} =& -\tilde{\chi}_{2,1,1}+\Big[((\beta+1)(\alpha_{10}+1)+k_E-1)\tilde{\chi}_{2,1,3}\\
&+\Big((\beta+1)(\tilde{\lambda}_2+\mathcal{R}+2)/2+k_E-1\Big)\tilde{\chi}_{2,1,2}\\
&+(\beta+1)\mathcal{R}\tilde{\chi}_{2,1,1}-(\beta+1)\tilde{\chi}_{2,2,4}+i\delta\eta\tilde{\omega}_3\Big]\cdot\\
&\cdot(1-k_E-(\beta+1)(\tilde{\lambda}_1+1)/2)^{-1}, \quad \text{(B36)}
\end{aligned}
$$

and

$$
\begin{aligned}
\tilde{R}_{1,1,1} =& -\frac{1}{2}i\alpha_{10}\mathcal{R}^2\tilde{A}_1(\tilde{\lambda}_2-\mathcal{R}),\\
\tilde{R}_{1,1,2} =& i\alpha_{10}\mathcal{R}^3\tilde{A}_2,\\
\tilde{R}_{1,1,3} =& \frac{1}{2}\mathcal{R}(\tilde{\lambda}_2+\mathcal{R})\Big[i\mathcal{R}\tilde{\omega}_1\tilde{J}_{1,1,1}-2i\tilde{\omega}_1\tilde{J}_{1,2,1}\\
&+\left(2\alpha_{10}\alpha_{11}+i\mathcal{R}\tilde{\omega}_2\right)\tilde{L}_{1,1}\Big]+2i\mathcal{R}\tilde{\omega}_1\tilde{J}_{1,3,1}\\
&-\tilde{L}_{2,1}\Big(\mathcal{R}(2\alpha_{10}\alpha_{11}+i\mathcal{R}\tilde{\omega}_2)+\tilde{\lambda}_2(4\alpha_{10}\alpha_{11}\\
&+i\mathcal{R}\tilde{\omega}_2)\Big)+\frac{1}{2}i\mathcal{R}\tilde{A}_1\Big(\alpha_{10}(\mathcal{R}^2-4\alpha_{11}\tilde{\omega}_1)\\
&+\mathcal{R}\tilde{\lambda}_2\left(\tilde{\omega}_3+\alpha_{10}\right)+\mathcal{R}^2\tilde{\omega}_3\Big),\\
\tilde{R}_{1,2,3} =& \frac{1}{2}\mathcal{R}(\tilde{\lambda}_2+\mathcal{R})\Big[i\mathcal{R}\tilde{\omega}_1\tilde{J}_{1,2,1}-4i\tilde{\omega}_1\tilde{J}_{1,3,1}\\
&+(2\alpha_{10}\alpha_{11}+i\mathcal{R}\tilde{\omega}_2)\tilde{L}_{2,1}\Big],\\
\tilde{R}_{1,3,3} =& \frac{1}{2}i\mathcal{R}^2\tilde{\omega}_1(\tilde{\lambda}_2+\mathcal{R})\tilde{J}_{1,3,1},\\
\tilde{R}_{1,1,4} =& -2i\alpha_{10}\mathcal{R}\tilde{\omega}_1\tilde{J}_{1,2,2}+2i\mathcal{R}\tilde{\omega}_1\tilde{J}_{1,3,2}\\
&+2\alpha_{10}\tilde{L}_{2,2}(\mathcal{R}(\alpha_{11}-i\tilde{\omega}_2)-4\alpha_{10}\alpha_{11})\\
&-2\alpha_{11}\alpha_{10}^2\mathcal{R}\tilde{L}_{1,2}-2i\alpha_{11}\alpha_{10}\mathcal{R}\tilde{A}_2\tilde{\omega}_1,\\
\tilde{R}_{1,2,4} =& -2\alpha_{11}\alpha_{10}^2\mathcal{R}\tilde{L}_{2,2}-4i\alpha_{10}\mathcal{R}\tilde{\omega}_1\tilde{J}_{1,3,2},\\
\tilde{R}_{2,1,1} =& \delta\tilde{A}_1, \quad \tilde{R}_{2,1,2}=\delta\tilde{A}_2, \quad \tilde{R}_{2,1,3}=-i\alpha_{10}\tilde{A}_3,\\
\tilde{R}_{2,1,4} =& i\tilde{\omega}_1\tilde{J}_{2,1,1}+\tilde{P}_{1,1}\left(2\alpha_{10}\alpha_{11}+i\tilde{\omega}_2\right)\\
&+i\tilde{A}_3\left(\tilde{\omega}_3+\alpha_{10}\right),\\
\tilde{R}_{2,2,4} =& i\tilde{\omega}_1\tilde{J}_{2,2,1}+\tilde{P}_{2,1}\left(2\alpha_{10}\alpha_{11}+i\tilde{\omega}_2\right),\\
\tilde{R}_{2,3,4} =& i\tilde{\omega}_1\tilde{J}_{2,3,1}. \quad \text{(B37)}
\end{aligned}
$$

From this, we deduce the relation

$$
\tilde{m}_3 \;=\; -\frac{i\beta\delta\mathcal{L}\tilde{\omega}_3}{(\beta+1)\left(1-k_E\right)}-i\mathcal{U}\tilde{\omega}_3
$$

$$+\frac{\mathcal{L}\left(\beta+k_E\right)+1-k_E}{\left(\beta+1\right)\left(1-k_E\right)\eta}\sum_{n=1}^{4}\tilde{\chi}_{2,1,n}. \qquad \text{(B38)}$$

for $\tilde{m}_3$. Requiring that $\Im(\tilde{m}_3)=0$, as expected on physical grounds, yields a condition for $\tilde{\omega}_1$ (the prefactor of $\tilde{\omega}_3$ vanishes by definition of the wavenumber α_{10} specific to the singular root). Specifically, we obtain the cubic equation (68) for $\tilde{\omega}_1$. The coefficients γ_j for $j=1,2,3$ that appear in (68) are given by

$$\begin{aligned}\gamma_1 =& \ \beta\left(8k_E+3\right)\left(2k_E^2+k_E+1\right)k_E^{-3}\left(2k_E+1\right)^{-1}\\ &-(2k_E+1)k_E^{-2}+\mathcal{O}(\beta^2,\mathcal{U}),\\ \gamma_2 =& -2\beta^{1/2}(2k_E+1)^{1/2}k_E^{-3/2}+\mathcal{O}(\beta^2,\mathcal{U}),\\ \gamma_3 =& \ \beta\mathcal{R}(2k_E\mathcal{R}+2k_E+\mathcal{R}+1)^{-1}+\mathcal{O}(\beta^2,\mathcal{U}). \qquad \text{(B39)}\end{aligned}$$

5. Fourth order in $\mathcal{V}^{1/3}$

The governing equations become

$$\begin{aligned}&\tilde{w}_{14}{}^{(4)}(z)+\mathcal{R}\tilde{w}_{14}{}^{(3)}(z)-2\alpha_{10}^2\tilde{w}_{14}''(z)-\alpha_{10}^2\mathcal{R}\tilde{w}_{14}'(z)\\ &+\alpha_{10}^4\tilde{w}_{14}(z)=i\mathcal{R}\tilde{\omega}_1\tilde{w}_{13}''(z)-i\alpha_{10}^2\mathcal{R}\tilde{\omega}_1\tilde{w}_{13}(z)\\ &+\left(2\alpha_{11}^2+i\mathcal{R}\tilde{\omega}_4\right)\tilde{w}_{10}''(z)+\alpha_{11}^2\mathcal{R}\tilde{w}_{10}'(z)\\ &-\alpha_{10}\left(2i\alpha_{11}\mathcal{R}\tilde{\omega}_2+i\alpha_{10}\mathcal{R}\tilde{\omega}_4+6\alpha_{10}\alpha_{11}^2\right)\tilde{w}_{10}(z)\\ &+i\mathcal{R}\left(\tilde{\omega}_3+\alpha_{10}-\alpha_{10}e^{-z\mathcal{R}}\right)\tilde{w}_{11}''(z)+i\alpha_{10}\mathcal{R}((\alpha_{10}^2\\ &+\mathcal{R}^2)e^{-z\mathcal{R}}-(2\alpha_{11}\tilde{\omega}_1+\alpha_{10}(\tilde{\omega}_3+\alpha_{10})))\tilde{w}_{11}(z)\\ &+2\alpha_{11}\alpha_{10}\mathcal{R}\tilde{w}_{12}'(z)+\left(4\alpha_{10}\alpha_{11}+i\mathcal{R}\tilde{\omega}_2\right)\tilde{w}_{12}''(z)\\ &-\alpha_{10}^2\left(4\alpha_{10}\alpha_{11}+i\mathcal{R}\tilde{\omega}_2\right)\tilde{w}_{12}(z), \qquad \text{(B40)}\end{aligned}$$

and

$$\begin{aligned}&\tilde{c}_{114}''(z)+\tilde{c}_{114}'(z)-\alpha_{10}^2\tilde{c}_{114}(z)=i\tilde{\omega}_1\tilde{c}_{113}(z)\\ &+\left(2\alpha_{10}\alpha_{11}+i\tilde{\omega}_2\right)\tilde{c}_{112}(z)\\ &+i\left(\tilde{\omega}_3+\alpha_{10}-\alpha_{10}e^{-z\mathcal{R}}\right)\tilde{c}_{111}(z)\\ &+\left(\alpha_{11}^2+i\tilde{\omega}_4\right)\tilde{c}_{110}(z)+\delta e^{-z}\tilde{w}_{11}(z), \qquad \text{(B41)}\end{aligned}$$

subject to

$$\tilde{w}_{14}(0)=0,\quad \tilde{w}_{14}'(0)=0,$$

$$\tilde{w}_{14}(z),\tilde{w}_{14}'(z),\tilde{c}_{114}(z)\to 0\quad \text{as}\quad z\to\infty,$$

$$\left(1-k_E\right)\tilde{c}_{114}(0)+(\beta+1)\tilde{c}_{114}'(0)-i\delta\eta\tilde{\omega}_4=0, \qquad \text{(B42)}$$

$$\begin{aligned}&\tilde{c}_{114}(0)\left(\mathcal{L}(k_E+\beta)+1-k_E\right)/\eta=i\beta\delta\mathcal{L}\tilde{\omega}_4\\ &+\left(\beta+1\right)\left(1-k_E\right)\left(\tilde{m}_4+\alpha_{11}^2\Gamma+i\mathcal{U}\tilde{\omega}_4\right). \qquad \text{(B43)}\end{aligned}$$

The solution is of the form

$$\begin{aligned}\tilde{w}_{14}(z)=&\sum_{n=1}^{5}\tilde{\phi}_{1,n,1}z^{n-1}e^{-\frac{1}{2}z\left(\tilde{\lambda}_2+\mathcal{R}\right)}+\\ &+\sum_{n=1}^{2}\tilde{\phi}_{1,n,2}z^{n-1}e^{-\frac{1}{2}z\left(\tilde{\lambda}_2+3\mathcal{R}\right)}\\ &+\sum_{n=1}^{4}\tilde{\phi}_{1,n,3}z^{n-1}e^{-\alpha_{10}z}+\\ &+\sum_{n=1}^{2}\tilde{\phi}_{1,n,4}z^{n-1}e^{-z\left(\alpha_{10}+\mathcal{R}\right)}, \qquad \text{(B44)}\end{aligned}$$

$$\begin{aligned}\tilde{c}_{114}(z)=&\sum_{n=1}^{5}\tilde{\phi}_{2,n,1}z^{n-1}e^{-\frac{1}{2}z\left(\tilde{\lambda}_1+1\right)}\\ &+\sum_{n=1}^{2}\tilde{\phi}_{2,n,2}z^{n-1}e^{-\frac{1}{2}z\left(\tilde{\lambda}_1+2\mathcal{R}+1\right)}\\ &+\sum_{n=1}^{2}\tilde{\phi}_{2,n,3}z^{n-1}e^{-\frac{1}{2}z\left(\tilde{\lambda}_2+\mathcal{R}+2\right)}\\ &+\sum_{n=1}^{2}\tilde{\phi}_{2,n,4}z^{n-1}e^{-(\alpha_{10}+1)z}, \qquad \text{(B45)}\end{aligned}$$

where the coefficients $\tilde{\phi}_{i,j,k}$ are given in Appendix C. Equation (B43) restricts the morphological number to satisfy

$$\tilde{m}_4=-\Gamma\alpha_{11}^2+\frac{\left(\beta+k_E\right)^2\Gamma_s}{(\beta+1)\eta k_E}\sum_{j=1}^{4}\tilde{\phi}_{2,1,j}, \qquad \text{(B46)}$$

which depends on ω_3 through $\tilde{\phi}_{2,1,j}$. Precisely, the form of $\tilde{m}_4$ is given by

$$\tilde{m}_4=\rho_1\tilde{\omega}_1^4+\rho_2\alpha_{11}\tilde{\omega}_1^2+\rho_3\tilde{\omega}_1+\rho_4\alpha_{11}^2+\rho_5\tilde{\omega}_3\tilde{\omega}_1, \quad \text{(B47)}$$

where $\rho_1,\ldots,\rho_5$ are numerical constants that depend on α_{10} and the physical parameters. To determine $\tilde{\omega}_3$, it is necessary to consider the next highest order.

6. Fifth order in $\mathcal{V}^{1/3}$

The governing equations at $\mathcal{O}(\mathcal{V}^{1/3})$ are

$$\begin{aligned}&\tilde{w}_{15}^{(4)}(z)+\mathcal{R}\tilde{w}_{15}^{(3)}(z)-2\alpha_{10}^2\tilde{w}_{15}''(z)-\alpha_{10}^2\mathcal{R}\tilde{w}_{15}'(z)\\ &+\alpha_{10}^4\tilde{w}_{15}(z)=i\mathcal{R}\tilde{\omega}_1\left(\tilde{w}_{14}''(z)-\alpha_{10}^2\tilde{w}_{14}(z)\right)\\ &+\left(4\alpha_{10}\alpha_{11}+i\mathcal{R}\tilde{\omega}_2\right)\left(\tilde{w}_{13}''(z)-\alpha_{10}^2\tilde{w}_{13}(z)\right)\\ &+2\alpha_{11}\alpha_{10}\mathcal{R}\tilde{w}_{13}'(z)+i\mathcal{R}\left(\tilde{\omega}_3+\alpha_{10}-\alpha_{10}e^{-z\mathcal{R}}\right)\cdot\\ &\cdot\left(\tilde{w}_{12}''(z)-\alpha_{10}^2\tilde{w}_{12}(z)\right)+\alpha_{11}^2\mathcal{R}\tilde{w}_{11}'(z)\\ &-i\alpha_{10}\mathcal{R}\left(2\alpha_{11}\tilde{\omega}_1-\mathcal{R}^2e^{-z\mathcal{R}}\right)\tilde{w}_{12}(z)\\ &+\left(2\alpha_{11}^2+i\mathcal{R}\tilde{\omega}_4\right)\tilde{w}_{11}''(z)-\alpha_{10}(2i\alpha_{11}\mathcal{R}\tilde{\omega}_2\\ &+i\alpha_{10}\mathcal{R}\tilde{\omega}_4+6\alpha_{10}\alpha_{11}^2)\tilde{w}_{11}(z)+i\mathcal{R}(\tilde{\omega}_5\\ &+\alpha_{11}-\alpha_{11}e^{-z\mathcal{R}})(\tilde{w}_{10}''(z)-3\alpha_{10}^2\tilde{w}_{10}(z))\\ &+i\mathcal{R}(2\alpha_{10}^2\tilde{\omega}_5-2\alpha_{11}\alpha_{10}\tilde{\omega}_3-\alpha_{11}^2\tilde{\omega}_1\end{aligned}$$

$$+ \alpha_{11}\mathcal{R}^2 e^{-z\mathcal{R}})\tilde{w}_{10}(z), \tag{B48}$$

$$\begin{aligned}\tilde{c}_{l15}''(z) + \tilde{c}_{l15}'(z) - \alpha_{10}^2\tilde{c}_{l15}(z) = i\tilde{\omega}_1\tilde{c}_{l14}(z)\\ + (2\alpha_{10}\alpha_{11} + i\tilde{\omega}_2)\,\tilde{c}_{l13}(z) + i(\tilde{\omega}_3 + \alpha_{10}\\ - \alpha_{10}e^{-z\mathcal{R}})\tilde{c}_{l12}(z) + \left(\alpha_{11}^2 + i\tilde{\omega}_4\right)\tilde{c}_{l11}(z)\\ + i\left(\tilde{\omega}_5 + \alpha_{11} - \alpha_{11}e^{-z\mathcal{R}}\right)\tilde{c}_{l10}(z)\\ + \delta e^{-z}\tilde{w}_{12}(z),\end{aligned} \tag{B49}$$

subject to

$$\tilde{w}_{15}(0) = 0, \quad \tilde{w}_{15}'(0) = 0,$$

$$\tilde{w}_{15}(z), \tilde{w}_{15}'(z), \tilde{c}_{l15}(z) \to 0 \quad \text{as} \quad z \to \infty,$$

$$(1 - k_E)\,\tilde{c}_{l15}(0) + (\beta+1)\tilde{c}_{l15}'(0) - i\delta\eta\tilde{\omega}_5 = 0, \tag{B50}$$

$$\begin{aligned}\tilde{c}_{l15}(0)\left(\mathcal{L}(k_E+\beta) + 1 - k_E\right)/\eta = i\beta\delta\mathcal{L}\tilde{\omega}_5\\ + (\beta+1)(1-k_E)(\tilde{m}_5 + i\mathcal{U}\tilde{\omega}_5).\end{aligned} \tag{B51}$$

The solution is of the form

$$\begin{aligned}\tilde{w}_{15}(z) = &\sum_{n=1}^{6}\tilde{\theta}_{1,n,1}z^{n-1}e^{-\frac{1}{2}z\left(\tilde{\lambda}_2+\mathcal{R}\right)}\\ &+ \sum_{n=3}^{6}\tilde{\theta}_{1,n,2}z^{n-1}e^{-\frac{1}{2}z\left(\tilde{\lambda}_2+3\mathcal{R}\right)}\\ &+ \sum_{n=1}^{5}\tilde{\theta}_{1,n,3}z^{n-1}e^{-\alpha_{10}z}\\ &+ \sum_{n=1}^{3}\tilde{\theta}_{1,n,4}z^{n-1}e^{-z(\alpha_{10}+\mathcal{R})},\end{aligned} \tag{B52}$$

$$\begin{aligned}\tilde{c}_{l15}(z) = &\sum_{n=1}^{6}\tilde{\theta}_{2,n,1}z^{n-1}e^{-\frac{1}{2}z\left(\tilde{\lambda}_1+1\right)}\\ &+ \sum_{n=1}^{3}\tilde{\theta}_{2,n,2}z^{n-1}e^{-\frac{1}{2}z\left(\tilde{\lambda}_1+2\mathcal{R}+1\right)}\\ &+ \sum_{n=1}^{3}\tilde{\theta}_{2,n,3}z^{n-1}e^{-\frac{1}{2}z\left(\tilde{\lambda}_2+\mathcal{R}+2\right)}\\ &+ \sum_{n=1}^{3}\tilde{\theta}_{2,n,4}z^{n-1}e^{-(\alpha_{10}+1)z},\end{aligned} \tag{B53}$$

where the coefficients $\tilde{\theta}_{i,j,k}$ are given in Appendix C. The relation (B51) gives that $\tilde{\omega}_3$ must satisfy

$$\begin{aligned}\tilde{m}_5 = &\frac{(\beta+k_E)^2\,\Gamma_s}{(\beta+1)\eta k_E}\sum_{j=1}^{4}\tilde{\theta}_{2,1,j}\\ &+ \frac{i\beta\delta\mathcal{L}\tilde{\omega}_5}{(\beta+1)(k_E-1)} - i\mathcal{U}\tilde{\omega}_5.\end{aligned} \tag{B54}$$

Requiring the morphological number to be real yields that $\tilde{m}_5 = 0$ and noting that $\alpha_1 = \alpha_{10}$ corresponds to the triple root eliminates $\tilde{\omega}_5$. The equation for $\tilde{\omega}_3$ then reduces to the form (70), where $\rho_6, \ldots, \rho_{12}$ are numerical constants that depend on the parameters. With $\tilde{\omega}_5$ known, the relation (B47) is sufficient to determine $\tilde{m}_4$, and hence the inverse morphological number is known to the required order in the inner region.

Appendix C

The coefficients $\tilde{\phi}_{i,j,k}$ are given by

$$\begin{aligned}\tilde{\phi}_{1,2,1} =&\mathcal{R}^{-4}\tilde{\lambda}_3^{-4}\Big[2\mathcal{R}^2\tilde{\lambda}_3^2(4\alpha_{10}^2 - 3\tilde{\lambda}_3)\tilde{g}_{1,2,3} + 16\mathcal{R}\tilde{\lambda}_3\cdot\\ &\cdot(-\tilde{\lambda}_3^2 - \mathcal{R}^2\tilde{\lambda}_3 + \alpha_{10}^2\mathcal{R}^2)\tilde{g}_{1,3,3} + 24(-40\alpha_{10}^4\tilde{\lambda}_3\\ &+ 15\alpha_{10}^2\tilde{\lambda}_3^2 - 5\tilde{\lambda}_3^3 - 4\alpha_{10}^4\left(4\alpha_{10}^2 + 3\mathcal{R}^2\right))\tilde{g}_{1,4,3}\\ &- 2\mathcal{R}^3\tilde{\lambda}_3^3\tilde{g}_{1,1,3}\Big],\\ \tilde{\phi}_{1,3,1} =&\mathcal{R}^{-3}\tilde{\lambda}_3^{-3}\Big[12(8\alpha_{10}^2\tilde{\lambda}_3 - 3\tilde{\lambda}_3^2 - 2\alpha_{10}^2(8\alpha_{10}^2 + \mathcal{R}^2))\cdot\\ &\cdot\tilde{g}_{1,4,3} - \mathcal{R}^2\tilde{\lambda}_3^2\tilde{g}_{1,2,3} + 2\mathcal{R}\tilde{\lambda}_3(4\alpha_{10}^2 - 3\tilde{\lambda}_3)\tilde{g}_{1,3,3}\Big],\\ \tilde{\phi}_{1,4,1} =&\Big[\tilde{g}_{1,4,3}(24\alpha_{10}^2 - 18\tilde{\lambda}_3) - 2\mathcal{R}\tilde{\lambda}_3\tilde{g}_{1,3,3}\Big](3\mathcal{R}^2\tilde{\lambda}_3^2)^{-1},\\ \tilde{\phi}_{1,5,1} =&-\tilde{g}_{1,4,3}(2\mathcal{R}\tilde{\lambda}_3)^{-1},\\ \tilde{\phi}_{1,1,2} =&2\tilde{g}_{1,1,1}(\mathcal{R}^2(\tilde{\lambda}_2 + \mathcal{R})(3\tilde{\lambda}_2 + 5\mathcal{R}))^{-1}\\ &+ 2\tilde{g}_{1,2,1}\left(19\mathcal{R}\tilde{\lambda}_2 + 20\alpha_{10}^2 + 21\mathcal{R}^2\right)\cdot\\ &\cdot(\mathcal{R}^3(\tilde{\lambda}_2 + \mathcal{R})^2(3\tilde{\lambda}_2 + 5\mathcal{R})^2)^{-1},\\ \tilde{\phi}_{1,2,2} =&2\tilde{g}_{1,2,1}(\mathcal{R}^2(\tilde{\lambda}_2 + \mathcal{R})(3\tilde{\lambda}_2 + 5\mathcal{R}))^{-1},\\ \tilde{\phi}_{1,2,3} =&(16\alpha_{10}^2 + 7\mathcal{R}^2 - 16\alpha_{10}\mathcal{R})\tilde{g}_{1,3,4}(4\alpha_{10}^4\mathcal{R}^3)^{-1}\\ &+ \Big[2\alpha_{10}\mathcal{R}\tilde{g}_{1,1,4} - 4\alpha_{10}\tilde{g}_{1,2,4} + 3\mathcal{R}\tilde{g}_{1,2,4}\Big]\cdot\\ &\cdot(4\alpha_{10}^3\mathcal{R}^2)^{-1},\\ \tilde{\phi}_{1,3,3} =&(\alpha_{10}\mathcal{R}\tilde{g}_{1,2,4} - 4\alpha_{10}\tilde{g}_{1,3,4} + 3\mathcal{R}\tilde{g}_{1,3,4})(4\alpha_{10}^3\mathcal{R}^2)^{-1},\\ \tilde{\phi}_{1,4,3} =&\tilde{g}_{1,3,4}(6\alpha_{10}^2\mathcal{R})^{-1},\\ \tilde{\phi}_{1,1,4} =&\tilde{g}_{1,1,2}(\alpha_{10}\mathcal{R}^2\,(2\alpha_{10} + \mathcal{R}))^{-1} + (6\alpha_{10}\mathcal{R} + 6\alpha_{10}^2\\ &+ \mathcal{R}^2)\tilde{g}_{1,2,2}(\alpha_{10}^2\mathcal{R}^3\,(2\alpha_{10} + \mathcal{R})^2)^{-1},\\ \tilde{\phi}_{1,2,4} =&\tilde{g}_{1,2,2}(\alpha_{10}\mathcal{R}^2\,(2\alpha_{10} + \mathcal{R}))^{-1},\\ \tilde{\phi}_{2,2,1} =&-(4\alpha_{10}^2\tilde{g}_{2,1,4} + \tilde{\lambda}_1\tilde{g}_{2,2,4} + \tilde{g}_{2,1,4} + 2\tilde{g}_{2,3,4})\tilde{\lambda}_1^{-3}\\ &- 6\tilde{g}_{2,4,4}\tilde{\lambda}_1^{-4},\\ \tilde{\phi}_{2,3,1} =&-(4\alpha_{10}^2\tilde{g}_{2,2,4} + 2\tilde{\lambda}_1\tilde{g}_{2,3,4} + \tilde{g}_{2,2,4} + 6\tilde{g}_{2,4,4})\cdot\\ &\cdot(2\tilde{\lambda}_1^3)^{-1},\\ \tilde{\phi}_{2,4,1} =&-3^{-1}\tilde{g}_{2,3,4}\tilde{\lambda}_1^{-1} - \tilde{g}_{2,4,4}\tilde{\lambda}_1^{-2},\\ \tilde{\phi}_{2,5,1} =&-4^{-1}\tilde{g}_{2,4,4}\tilde{\lambda}_1^{-1},\\ \tilde{\phi}_{2,1,2} =&\tilde{g}_{2,1,1}\mathcal{R}^{-1}(\tilde{\lambda}_1 + \mathcal{R})^{-1} + \tilde{g}_{2,2,1}(\tilde{\lambda}_1 + 2\mathcal{R})\cdot\end{aligned}$$

$$\cdot \mathcal{R}^{-2}(\tilde{\lambda}_1+\mathcal{R})^{-2},$$
$$\tilde{\phi}_{2,2,2} = \tilde{g}_{2,2,1}\mathcal{R}^{-1}(\tilde{\lambda}_1+\mathcal{R})^{-1},$$
$$\tilde{\phi}_{2,1,3} = 2\tilde{g}_{2,1,2}(\mathcal{R}+1)^{-1}(\tilde{\lambda}_2+\mathcal{R})^{-1}+4\tilde{g}_{2,2,2}(\tilde{\lambda}_2 + \mathcal{R}+1)(\mathcal{R}+1)^{-2}(\tilde{\lambda}_2+\mathcal{R})^{-2},$$
$$\tilde{\phi}_{2,2,3} = 2\tilde{g}_{2,2,2}(\mathcal{R}+1)^{-1}(\tilde{\lambda}_2+\mathcal{R})^{-1},$$
$$\tilde{\phi}_{2,1,4} = (\alpha_{10}\tilde{g}_{2,1,3}+2\alpha_{10}\tilde{g}_{2,2,3}+\tilde{g}_{2,2,3})\alpha_{10}^{-2},$$
$$\tilde{\phi}_{2,2,4} = \tilde{g}_{2,2,3}\alpha_{10}^{-1}, \tag{C1}$$

and

$$\tilde{\phi}_{1,1,1} = 2\Big(\Sigma_{n=1}^{4}\tilde{\phi}_{1,2,n}-\mathcal{R}\tilde{\phi}_{1,1,2}-\mathcal{R}\tilde{\phi}_{1,1,4}\Big)\cdot \cdot(\tilde{\lambda}_2-2\alpha_{10}+\mathcal{R})^{-1}-\tilde{\phi}_{1,1,2}, \tag{C2}$$
$$\tilde{\phi}_{1,1,3} = -2\Big(\Sigma_{n=1}^{4}\tilde{\phi}_{1,2,n}-\mathcal{R}\tilde{\phi}_{1,1,2}-\mathcal{R}\tilde{\phi}_{1,1,4}\Big)\cdot \cdot(\tilde{\lambda}_2-2\alpha_{10}+\mathcal{R})^{-1}-\tilde{\phi}_{1,1,4}, \tag{C3}$$
$$\tilde{\phi}_{2,1,1} = \Big[-2(\beta+1)\left(\Sigma_{n=1}^{4}\tilde{\phi}_{2,2,n}-(\alpha_{10}+1)\tilde{\phi}_{2,1,4}\right) + 2\left(k_E-1\right)\Sigma_{n=2}^{4}\tilde{\phi}_{2,1,n}+(\beta+1)\tilde{\phi}_{2,1,2}(\tilde{\lambda}_1 + 2\mathcal{R}+1)+(\beta+1)\tilde{\phi}_{2,1,3}(\tilde{\lambda}_2+\mathcal{R}+2)+ + 2i\delta\eta\tilde{\omega}_4\Big]\Big[2(1-k_E)-(\beta+1)(\tilde{\lambda}_1+1)\Big]^{-1}. \tag{C4}$$

The $\tilde{g}_{i,j,k}$ are given by

$$\tilde{g}_{1,1,1} = i\alpha_{10}\mathcal{R}\tilde{L}_{2,1}(\tilde{\lambda}_2+\mathcal{R})-i\alpha_{10}\mathcal{R}^2\tilde{L}_{1,1}(\tilde{\lambda}_2-\mathcal{R})/2 + i\mathcal{R}^2\tilde{\omega}_1\tilde{\chi}_{1,1,1}(3\tilde{\lambda}_2+5\mathcal{R})/2,$$
$$\tilde{g}_{1,2,1} = -i\alpha_{10}\mathcal{R}^2\tilde{L}_{2,1}(\tilde{\lambda}_2-\mathcal{R})/2,$$
$$\tilde{g}_{1,1,2} = i\alpha_{10}\mathcal{R}^3\tilde{L}_{1,2}+2i\alpha_{10}^2\mathcal{R}\tilde{L}_{2,2} + i\mathcal{R}^2\tilde{\omega}_1\left(2\alpha_{10}+\mathcal{R}\right)\tilde{\chi}_{1,1,2},$$
$$\tilde{g}_{1,2,2} = i\alpha_{10}\mathcal{R}^3\tilde{L}_{2,2},$$
$$\tilde{g}_{1,1,3} = \alpha_{10}\alpha_{11}\mathcal{R}\tilde{J}_{1,1,1}(\tilde{\lambda}_2+\mathcal{R})-2\alpha_{10}\alpha_{11}\tilde{J}_{1,2,1}(2\tilde{\lambda}_2 + \mathcal{R})+8\alpha_{10}\alpha_{11}\tilde{J}_{1,3,1}+\frac{1}{2}i\mathcal{R}^2\tilde{\omega}_1\tilde{\chi}_{1,1,3}(\tilde{\lambda}_2 + \mathcal{R})-i\mathcal{R}\tilde{\omega}_1\tilde{\chi}_{1,2,3}(\tilde{\lambda}_2+\mathcal{R})+2i\mathcal{R}\tilde{\omega}_1\tilde{\chi}_{1,3,3} + \alpha_{11}^2\tilde{A}_1(\mathcal{R}\left(\tilde{\lambda}_2+\mathcal{R}\right)-8\alpha_{10}^2)/2+i\mathcal{R}\tilde{L}_{1,1}\cdot \cdot\left(\alpha_{10}\left(\mathcal{R}^2-4\alpha_{11}\tilde{\omega}_1\right)+\mathcal{R}\tilde{\lambda}_2\left(\tilde{\omega}_3+\alpha_{10}\right) + \mathcal{R}^2\tilde{\omega}_3\right)/2-i\mathcal{R}\tilde{L}_{2,1}\left(\tilde{\omega}_3+\alpha_{10}\right)\left(\tilde{\lambda}_2+\mathcal{R}\right),$$
$$\tilde{g}_{1,2,3} = \alpha_{10}\alpha_{11}\mathcal{R}\tilde{J}_{1,2,1}(\tilde{\lambda}_2+\mathcal{R})-4\alpha_{10}\alpha_{11}\tilde{J}_{1,3,1}(2\tilde{\lambda}_2 + \mathcal{R})+\frac{1}{2}i\mathcal{R}^2\tilde{\omega}_1\tilde{\chi}_{1,2,3}(\tilde{\lambda}_2+\mathcal{R})+6i\mathcal{R}\tilde{\omega}_1\tilde{\chi}_{1,4,3} - 2i\mathcal{R}\tilde{\omega}_1\tilde{\chi}_{1,3,3}(\tilde{\lambda}_2+\mathcal{R})+i\mathcal{R}\tilde{L}_{2,1}(\alpha_{10}(\mathcal{R}^2 - 4\alpha_{11}\tilde{\omega}_1)+\mathcal{R}\tilde{\lambda}_2(\tilde{\omega}_3+\alpha_{10})+\mathcal{R}^2\tilde{\omega}_3)/2,$$
$$\tilde{g}_{1,3,3} = \mathcal{R}\tilde{J}_{1,3,1}(\tilde{\lambda}_2+\mathcal{R})(2\alpha_{10}\alpha_{11}+i\mathcal{R}\tilde{\omega}_2)/2+i\mathcal{R}^2\cdot \cdot\tilde{\omega}_1\tilde{\chi}_{1,3,3}(\tilde{\lambda}_2+\mathcal{R})/2-3i\mathcal{R}\tilde{\omega}_1\tilde{\chi}_{1,4,3}(\tilde{\lambda}_2+\mathcal{R}),$$
$$\tilde{g}_{1,4,3} = i\mathcal{R}^2\tilde{\omega}_1\tilde{\chi}_{1,4,3}(\tilde{\lambda}_2+\mathcal{R})/2,$$
$$\tilde{g}_{1,1,4} = -2\alpha_{11}\alpha_{10}^2\mathcal{R}\tilde{J}_{1,1,2}+2\alpha_{11}\alpha_{10}\left(\mathcal{R}-4\alpha_{10}\right)\tilde{J}_{1,2,2} + 8\alpha_{11}\alpha_{10}\tilde{J}_{1,3,2}-2i\alpha_{11}\alpha_{10}\mathcal{R}\tilde{\omega}_1\tilde{L}_{1,2} - 2i\alpha_{10}\mathcal{R}\tilde{L}_{2,2}\left(\tilde{\omega}_3+\alpha_{10}\right)-2i\alpha_{10}\mathcal{R}\tilde{\omega}_1\tilde{\chi}_{1,2,4} + 2i\mathcal{R}\tilde{\omega}_1\tilde{\chi}_{1,3,4}-\alpha_{10}\alpha_{11}^2\tilde{A}_2\left(4\alpha_{10}+\mathcal{R}\right),$$
$$\tilde{g}_{1,2,4} = -2\alpha_{11}\alpha_{10}^2\mathcal{R}\tilde{J}_{1,2,2}+4\alpha_{11}\alpha_{10}\left(\mathcal{R}-4\alpha_{10}\right)\tilde{J}_{1,3,2} - 2i\alpha_{11}\alpha_{10}\mathcal{R}\tilde{\omega}_1\tilde{L}_{2,2}-4i\alpha_{10}\mathcal{R}\tilde{\omega}_1\tilde{\chi}_{1,3,4} + 6i\mathcal{R}\tilde{\omega}_1\tilde{\chi}_{1,4,4},$$
$$\tilde{g}_{1,3,4} = -2\alpha_{11}\alpha_{10}^2\mathcal{R}\tilde{J}_{1,3,2}-6i\alpha_{10}\mathcal{R}\tilde{\omega}_1\tilde{\chi}_{1,4,4},$$
$$\tilde{g}_{2,1,1} = i\tilde{\omega}_1\tilde{\chi}_{2,1,1}-i\alpha_{10}\tilde{P}_{1,1},$$
$$\tilde{g}_{2,2,1} = -i\alpha_{10}\tilde{P}_{2,1},$$
$$\tilde{g}_{2,1,2} = \delta\tilde{L}_{1,1}+i\tilde{\omega}_1\tilde{\chi}_{2,1,2},$$
$$\tilde{g}_{2,2,2} = \delta\tilde{L}_{2,1},$$
$$\tilde{g}_{2,1,3} = \delta\tilde{L}_{1,2}+i\tilde{\omega}_1\tilde{\chi}_{2,1,3},$$
$$\tilde{g}_{2,2,3} = \delta\tilde{L}_{2,2},$$
$$\tilde{g}_{2,1,4} = \tilde{J}_{2,1,1}\left(2\alpha_{10}\alpha_{11}+i\tilde{\omega}_2\right)+i\tilde{P}_{1,1}\left(\tilde{\omega}_3+\alpha_{10}\right) + i\tilde{\omega}_1\tilde{\chi}_{2,1,4}+\tilde{A}_3\alpha_{11}^2,$$
$$\tilde{g}_{2,2,4} = \tilde{J}_{2,2,1}\left(2\alpha_{10}\alpha_{11}+i\tilde{\omega}_2\right)+i\tilde{P}_{2,1}\left(\tilde{\omega}_3+\alpha_{10}\right) + i\tilde{\omega}_1\tilde{\chi}_{2,2,4},$$
$$\tilde{g}_{2,3,4} = \tilde{J}_{2,3,1}\left(2\alpha_{10}\alpha_{11}+i\tilde{\omega}_2\right)+i\tilde{\omega}_1\tilde{\chi}_{2,3,4},$$
$$\tilde{g}_{2,4,4} = i\tilde{\omega}_1\tilde{\chi}_{2,4,4} \tag{C5}$$

The coefficients $\tilde{\theta}_{i,j,k}$ are given by

$$\tilde{\theta}_{1,2,1} = \Big[-16\mathcal{R}^2(4\alpha_{10}^2+\mathcal{R}^2)\tilde{f}_{1,3,3}(24\alpha_{10}^4\tilde{\lambda}_3-13\alpha_{10}^2\tilde{\lambda}_3^2 + 3\tilde{\lambda}_3^3-16\alpha_{10}^6)-24\mathcal{R}\tilde{f}_{1,4,3}(-32\alpha_{10}^6\tilde{\lambda}_3 + 46\alpha_{10}^4\tilde{\lambda}_3^2-15\alpha_{10}^2\tilde{\lambda}_3^3+5\tilde{\lambda}_3^4+24\alpha_{10}^6(4\alpha_{10}^2 + \mathcal{R}^2))-8\mathcal{R}^4(4\alpha_{10}^2+\mathcal{R}^2)^2\tilde{f}_{1,1,3}(-4\alpha_{10}^2\tilde{\lambda}_3 + \tilde{\lambda}_3^2+4\alpha_{10}^4)-48\tilde{f}_{1,5,3}(-384\alpha_{10}^6\tilde{\lambda}_3+252\alpha_{10}^4\tilde{\lambda}_3^2 - 50\alpha_{10}^2\tilde{\lambda}_3^3+15\tilde{\lambda}_3^4+16\alpha_{10}^4(15\alpha_{10}^2\mathcal{R}^2+60\alpha_{10}^4 + \mathcal{R}^4))-4\mathcal{R}^3\tilde{\lambda}_3(4\alpha_{10}^2+\mathcal{R}^2)\tilde{f}_{1,2,3}(-10\alpha_{10}^2\tilde{\lambda}_3 + 3\tilde{\lambda}_3^2+8\alpha_{10}^4)\Big](\mathcal{R}^5\tilde{\lambda}_3^5)^{-1}, \tag{C6}$$

$$\tilde{\theta}_{1,3,1} = \Big[4\mathcal{R}^2(4\alpha_{10}^2+\mathcal{R}^2)\tilde{f}_{1,3,3}(10\alpha_{10}^2\tilde{\lambda}_3-3\tilde{\lambda}_3^2-8\alpha_{10}^4) - 12\mathcal{R}\tilde{f}_{1,4,3}(8\alpha_{10}^4\tilde{\lambda}_3-7\alpha_{10}^2\tilde{\lambda}_3^2+3\tilde{\lambda}_3^3+4\alpha_{10}^4\cdot \cdot(4\alpha_{10}^2+\mathcal{R}^2))-48\tilde{f}_{1,5,3}(40\alpha_{10}^4\tilde{\lambda}_3-15\alpha_{10}^2\tilde{\lambda}_3^2 + 5\tilde{\lambda}_3^3+4\alpha_{10}^4(4\alpha_{10}^2+3\mathcal{R}^2))-2\mathcal{R}^3\tilde{\lambda}_3(4\alpha_{10}^2 + \mathcal{R}^2)\tilde{f}_{1,2,3}(\mathcal{R}\tilde{\lambda}_2+2\alpha_{10}^2+\mathcal{R}^2)\Big](\mathcal{R}^4\tilde{\lambda}_3^4)^{-1}, \tag{C7}$$

$$\tilde{\theta}_{1,4,1} = \Big[-4\mathcal{R}^2(4\alpha_{10}^2+\mathcal{R}^2)\tilde{f}_{1,3,3}(\mathcal{R}\tilde{\lambda}_2+2\alpha_{10}^2+\mathcal{R}^2) - 48\tilde{f}_{1,5,3}(3\tilde{\lambda}_3^2-8\alpha_{10}^2\tilde{\lambda}_3+2\alpha_{10}^2(8\alpha_{10}^2+\mathcal{R}^2))$$

$$- 6\mathcal{R}\tilde{\lambda}_3\tilde{f}_{1,4,3}(3\tilde{\lambda}_3 - 4\alpha_{10}^2)\Big](3\mathcal{R}^3\tilde{\lambda}_3^3)^{-1}, \tag{C8}$$

$$\tilde{\theta}_{1,5,1} = -(\mathcal{R}\tilde{f}_{1,4,3}\tilde{\lambda}_3 + 4\tilde{f}_{1,5,3}(3\mathcal{R}(\tilde{\lambda}_2 + \mathcal{R}) + 8\alpha_{10}^2))\cdot \cdot (2\mathcal{R}^2\tilde{\lambda}_3^2)^{-1}, \tag{C9}$$

$$\tilde{\theta}_{1,6,1} = -2\tilde{f}_{1,5,3}(5\mathcal{R}\tilde{\lambda}_3)^{-1}, \tag{C10}$$

$$\tilde{\theta}_{1,1,2} = \tilde{f}_{1,1,1}(2\mathcal{R}^2(2\mathcal{R}\tilde{\lambda}_2 + 3\alpha_{10}^2 + 2\mathcal{R}^2))^{-1} + \tilde{f}_{1,2,1}(19\mathcal{R}\tilde{\lambda}_2 + 20\alpha_{10}^2 + 21\mathcal{R}^2)(8\mathcal{R}^3(2\mathcal{R}\tilde{\lambda}_2 + 3\alpha_{10}^2 + 2\mathcal{R}^2)^2)^{-1} + \tilde{f}_{1,3,1}(258\alpha_{10}^2\mathcal{R}\tilde{\lambda}_2 + 255\mathcal{R}^3\tilde{\lambda}_2 + 744\alpha_{10}^2\mathcal{R}^2 + 152\alpha_{10}^4 + 257\mathcal{R}^4)\cdot \cdot (8\mathcal{R}^4(2\mathcal{R}\tilde{\lambda}_2 + 3\alpha_{10}^2 + 2\mathcal{R}^2)^3)^{-1}, \tag{C11}$$

$$\tilde{\theta}_{1,2,2} = \tilde{f}_{1,2,1}(2\mathcal{R}^2(2\mathcal{R}\tilde{\lambda}_2 + 3\alpha_{10}^2 + 2\mathcal{R}^2))^{-1} + \tilde{f}_{1,3,1}\cdot \cdot (19\mathcal{R}\tilde{\lambda}_2 + 20\alpha_{10}^2 + 21\mathcal{R}^2)(4\mathcal{R}^3(2\mathcal{R}\tilde{\lambda}_2 + 3\alpha_{10}^2 + 2\mathcal{R}^2)^2)^{-1}, \tag{C12}$$

$$\tilde{\theta}_{1,3,2} = \tilde{f}_{1,3,1}(2\mathcal{R}^2(2\mathcal{R}\tilde{\lambda}_2 + 3\alpha_{10}^2 + 2\mathcal{R}^2))^{-1}, \tag{C13}$$

$$\tilde{\theta}_{1,2,3} = (2\alpha_{10}\mathcal{R}\tilde{f}_{1,1,4} - 4\alpha_{10}\tilde{f}_{1,2,4} + 3\mathcal{R}\tilde{f}_{1,2,4})\cdot \cdot (4\alpha_{10}^3\mathcal{R}^2)^{-1} + \tilde{f}_{1,3,4}(16\alpha_{10}^2 + 7\mathcal{R}^2 - 16\alpha_{10}\mathcal{R})(4\alpha_{10}^4\mathcal{R}^3)^{-1} + 3\tilde{f}_{1,4,4}(80\alpha_{10}^2\mathcal{R} - 48\alpha_{10}\mathcal{R}^2 - 64\alpha_{10}^3 + 15\mathcal{R}^3)/(8\alpha_{10}^5\mathcal{R}^4), \tag{C14}$$

$$\tilde{\theta}_{1,3,3} = 3\tilde{f}_{1,4,4}(16\alpha_{10}^2 - 16\alpha_{10}\mathcal{R} + 7\mathcal{R}^2)(8\alpha_{10}^4\mathcal{R}^3)^{-1} + (\alpha_{10}\mathcal{R}\tilde{f}_{1,2,4} + 3\mathcal{R}\tilde{f}_{1,3,4} - 4\alpha_{10}\tilde{f}_{1,3,4})\cdot \cdot (4\alpha_{10}^3\mathcal{R}^2)^{-1}, \tag{C15}$$

$$\tilde{\theta}_{1,4,3} = (2\alpha_{10}\mathcal{R}\tilde{f}_{1,3,4} - 12\alpha_{10}\tilde{f}_{1,4,4} + 9\mathcal{R}\tilde{f}_{1,4,4})\cdot \cdot (12\alpha_{10}^3\mathcal{R}^2)^{-1}, \tag{C16}$$

$$\tilde{\theta}_{1,5,3} = \tilde{f}_{1,4,4}(8\alpha_{10}^2\mathcal{R})^{-1}, \tag{C17}$$

$$\tilde{\theta}_{1,1,4} = \tilde{f}_{1,1,2}(\alpha_{10}\mathcal{R}^2(2\alpha_{10} + \mathcal{R}))^{-1} + (6\alpha_{10}\mathcal{R} + 6\alpha_{10}^2 + \mathcal{R}^2)\tilde{f}_{1,2,2}(\alpha_{10}^2\mathcal{R}^3\,(2\alpha_{10} + \mathcal{R})^2)^{-1} + 2(\mathcal{R}^4 + 9\alpha_{10}\mathcal{R}^3 + 33\alpha_{10}^2\mathcal{R}^2 + 50\alpha_{10}^3\mathcal{R} + 28\alpha_{10}^4)\cdot \cdot \tilde{f}_{1,3,2}(\alpha_{10}^3\mathcal{R}^4\,(2\alpha_{10} + \mathcal{R})^3)^{-1}, \tag{C18}$$

$$\tilde{\theta}_{1,2,4} = \tilde{f}_{1,2,2}(\alpha_{10}\mathcal{R}^2(2\alpha_{10} + \mathcal{R}))^{-1} + 2\tilde{f}_{1,3,2}(6\alpha_{10}\mathcal{R} + 6\alpha_{10}^2 + \mathcal{R}^2)(\alpha_{10}^2\mathcal{R}^3(2\alpha_{10} + \mathcal{R})^2)^{-1}, \tag{C19}$$

$$\tilde{\theta}_{1,3,4} = \tilde{f}_{1,3,2}(\alpha_{10}\mathcal{R}^2\,(2\alpha_{10} + \mathcal{R}))^{-1}, \tag{C20}$$

$$\tilde{\theta}_{2,2,1} = -\tilde{f}_{2,1,4}\tilde{\lambda}_1^{-1} - \tilde{f}_{2,2,4}\tilde{\lambda}_1^{-2} - 6\tilde{f}_{2,4,4}\tilde{\lambda}_1^{-4} - 2\tilde{f}_{2,3,4}\tilde{\lambda}_1^{-3} - 24\tilde{f}_{2,5,4}\tilde{\lambda}_1^{-5}, \tag{C21}$$

$$\tilde{\theta}_{2,3,1} = -\tilde{f}_{2,2,4}2\tilde{\lambda}_1^{-1} - \tilde{f}_{2,3,4}\tilde{\lambda}_1^{-2} - 12\tilde{f}_{2,5,4}\tilde{\lambda}_1^{-4} - 3\tilde{f}_{2,4,4}\tilde{\lambda}_1^{-3}, \tag{C22}$$

$$\tilde{\theta}_{2,4,1} = -\tilde{f}_{2,3,4}3\tilde{\lambda}_1^{-1} - \tilde{f}_{2,4,4}\tilde{\lambda}_1^{-2} - 4\tilde{f}_{2,5,4}\tilde{\lambda}_1^{-3}, \tag{C23}$$

$$\tilde{\theta}_{2,5,1} = -\tilde{f}_{2,4,4}4\tilde{\lambda}_1^{-1} - \tilde{f}_{2,5,4}\tilde{\lambda}_1^{-2}, \tag{C24}$$

$$\tilde{\theta}_{2,6,1} = -\tilde{f}_{2,5,4}5\tilde{\lambda}_1^{-2}, \tag{C25}$$

$$\tilde{\theta}_{2,1,2} = 2\tilde{f}_{2,3,1}(3\mathcal{R}\tilde{\lambda}_1 + 4\alpha_{10}^2 + 3\mathcal{R}^2 + 1)\cdot \cdot (\mathcal{R}^3(\tilde{\lambda}_1 + \mathcal{R})^3)^{-1} + \tilde{f}_{2,2,1}(\tilde{\lambda}_1 + 2\mathcal{R})(\mathcal{R}^2\cdot \cdot (\tilde{\lambda}_1 + \mathcal{R})^2)^{-1} + \tilde{f}_{2,1,1}(\mathcal{R}(\tilde{\lambda}_1 + \mathcal{R}))^{-1}, \tag{C26}$$

$$\tilde{\theta}_{2,2,2} = 2\tilde{f}_{2,3,1}(\tilde{\lambda}_1 + 2\mathcal{R})(\mathcal{R}^2(\tilde{\lambda}_1 + \mathcal{R})^2)^{-1} + \tilde{f}_{2,2,1}(\mathcal{R}(\tilde{\lambda}_1 + \mathcal{R}))^{-1}, \tag{C27}$$

$$\tilde{\theta}_{2,3,2} = \tilde{f}_{2,3,1}(\mathcal{R}(\tilde{\lambda}_1 + \mathcal{R}))^{-1}, \tag{C28}$$

$$\tilde{\theta}_{2,1,3} = 8\tilde{f}_{2,3,2}(3(\mathcal{R}+1)(\tilde{\lambda}_2 + \mathcal{R}) + 8\alpha_{10}^2 + 2)\cdot \cdot ((\mathcal{R}+1)^3(\tilde{\lambda}_2 + \mathcal{R})^3)^{-1} + 2\tilde{f}_{2,1,2}\cdot \cdot ((\mathcal{R}+1)(\tilde{\lambda}_2 + \mathcal{R}))^{-1} + 4\tilde{f}_{2,2,2}(\tilde{\lambda}_2 + \mathcal{R} + 1)((\mathcal{R}+1)^2(\tilde{\lambda}_2 + \mathcal{R})^2)^{-1}, \tag{C29}$$

$$\tilde{\theta}_{2,2,3} = 2\tilde{f}_{2,2,2}((\mathcal{R}+1)(\tilde{\lambda}_2 + \mathcal{R}))^{-1} + 8\tilde{f}_{2,3,2}(\tilde{\lambda}_2 + \mathcal{R} + 1)((\mathcal{R}+1)^2(\tilde{\lambda}_2 + \mathcal{R})^2)^{-1}, \tag{C30}$$

$$\tilde{\theta}_{2,3,3} = 2\tilde{f}_{2,3,2}((\mathcal{R}+1)(\tilde{\lambda}_2 + \mathcal{R}))^{-1}, \tag{C31}$$

$$\tilde{\theta}_{2,1,4} = \tilde{f}_{2,1,3}\alpha_{10}^{-1} + (\alpha_{10}^{-2} + 2\alpha_{10}^{-1})\tilde{f}_{2,2,3} + 2\tilde{f}_{2,3,3}(4\alpha_{10}^2 + 3\alpha_{10} + 1)\alpha_{10}^{-3}, \tag{C32}$$

$$\tilde{\theta}_{2,2,4} = \tilde{f}_{2,2,3}\alpha_{10}^{-1} + \tilde{f}_{2,3,3}(4\alpha_{10} + 2)\alpha_{10}^{-2}, \tag{C33}$$

$$\tilde{\theta}_{2,3,4} = \tilde{f}_{2,3,3}\alpha_{10}^{-1}, \tag{C34}$$

and

$$\tilde{\theta}_{1,1,1} = 2\left(\Sigma_{n=1}^4\tilde{\theta}_{1,2,n} - \mathcal{R}\tilde{\theta}_{1,1,2} - \mathcal{R}\tilde{\theta}_{1,1,4}\right)\cdot \cdot (\tilde{\lambda}_2 - 2\alpha_{10} + \mathcal{R})^{-1} - \tilde{\theta}_{1,1,2}, \tag{C35}$$

$$\tilde{\theta}_{1,1,3} = -2\left(\Sigma_{n=1}^4\tilde{\theta}_{1,2,n} - \mathcal{R}\tilde{\theta}_{1,1,2} - \mathcal{R}\tilde{\theta}_{1,1,4}\right)\cdot \cdot (\tilde{\lambda}_2 - 2\alpha_{10} + \mathcal{R})^{-1} - \tilde{\theta}_{1,1,4}, \tag{C36}$$

$$\tilde{\theta}_{2,1,1} = \Big[2(\beta+1)\Big(-\Sigma_{n=1}^4\tilde{\theta}_{2,2,n} + (\alpha_{10}+1)\,\tilde{\theta}_{2,1,4} + \mathcal{R}\tilde{\theta}_{2,1,2}\Big) + 2(k_E - 1)(\tilde{\theta}_{2,1,3} + \tilde{\theta}_{2,1,4}) + (\beta+1)\tilde{\theta}_{2,1,3}(\tilde{\lambda}_2 + \mathcal{R} + 2) + 2i\delta\eta\tilde{\omega}_5\Big]\cdot \cdot \Big[2(1 - k_E) - (\beta+1)(\tilde{\lambda}_1 + 1)\Big]^{-1} - \tilde{\theta}_{2,1,2}, \tag{C37}$$

where

$$\tilde{f}_{1,1,1} = -i\alpha_{10}\mathcal{R}^2\tilde{J}_{1,1,1}(\tilde{\lambda}_2 - \mathcal{R})/2 - 2i\alpha_{10}\mathcal{R}\tilde{J}_{1,3,1} + i\alpha_{10}\mathcal{R}\tilde{J}_{1,2,1}(\tilde{\lambda}_2 + \mathcal{R}) + \mathcal{R}\tilde{\chi}_{1,1,1}(\tilde{\lambda}_2\cdot \cdot (10\alpha_{10}\alpha_{11} + 3i\mathcal{R}\tilde{\omega}_2)/2 + \mathcal{R}(14\alpha_{10}\alpha_{11} + 5i\mathcal{R}\tilde{\omega}_2)) + \frac{1}{2}i\mathcal{R}^2\tilde{\omega}_1\tilde{\phi}_{1,1,2}(3\tilde{\lambda}_2 + 5\mathcal{R}) - i\mathcal{R}\tilde{\omega}_1\tilde{\phi}_{1,2,2}(\tilde{\lambda}_2 + 3\mathcal{R}) + i\alpha_{11}\mathcal{R}\tilde{A}_1(\mathcal{R}\cdot \cdot (\mathcal{R} - \tilde{\lambda}_2) + 4\alpha_{10}^2)/2, \tag{C38}$$

$$\tilde{f}_{1,2,1} = -i\alpha_{10}\mathcal{R}^2\tilde{J}_{1,2,1}(\tilde{\lambda}_2 - \mathcal{R})/2 + 2i\alpha_{10}\mathcal{R}\tilde{J}_{1,3,1}\cdot \cdot (\tilde{\lambda}_2 + \mathcal{R}) + i\mathcal{R}^2\tilde{\omega}_1\tilde{\phi}_{1,2,2}(3\tilde{\lambda}_2 + 5\mathcal{R})/2, \tag{C39}$$

$$\tilde{f}_{1,3,1} = -i\alpha_{10}\mathcal{R}^2\tilde{J}_{1,3,1}(\tilde{\lambda}_2 - \mathcal{R})/2, \tag{C40}$$

$$\tilde{f}_{1,1,2} = i\alpha_{10}\mathcal{R}^3\tilde{J}_{1,1,2} + 2i\alpha_{10}^2\mathcal{R}\tilde{J}_{1,2,2} - 2i\alpha_{10}\mathcal{R}\tilde{J}_{1,3,2} + i\mathcal{R}^2\tilde{\omega}_1\,(2\alpha_{10} + \mathcal{R})\,\tilde{\phi}_{1,1,4} + \mathcal{R}\tilde{\chi}_{1,1,2}(2\alpha_{10}\alpha_{11}\cdot \cdot (3\alpha_{10} + \mathcal{R}) + i\mathcal{R}\tilde{\omega}_2(2\alpha_{10} + \mathcal{R})) - 2i\mathcal{R}\tilde{\omega}_1\cdot$$

$$\cdot (\alpha_{10} + \mathcal{R})\tilde{\phi}_{1,2,4} + i\alpha_{11}\mathcal{R}\tilde{A}_2(2\alpha_{10}^2 + \mathcal{R}^2), \tag{C41}$$

$$\begin{aligned}\tilde{f}_{1,2,2} =& i\alpha_{10}\mathcal{R}^3\tilde{J}_{1,2,2} + 4i\alpha_{10}^2\mathcal{R}\tilde{J}_{1,3,2} \\ &+ i\mathcal{R}^2\tilde{\omega}_1\left(2\alpha_{10} + \mathcal{R}\right)\tilde{\phi}_{1,2,4},\end{aligned} \tag{C42}$$

$$\tilde{f}_{1,3,2} = i\alpha_{10}\mathcal{R}^3\tilde{J}_{1,3,2}, \tag{C43}$$

$$\begin{aligned}\tilde{f}_{1,1,3} =& i\mathcal{R}\tilde{J}_{1,1,1}(\alpha_{10}(\mathcal{R}^2 - 4\alpha_{11}\tilde{\omega}_1) + \mathcal{R}\tilde{\lambda}_2(\tilde{\omega}_3 + \alpha_{10}) + \\ &\mathcal{R}^2\tilde{\omega}_3)/2 - i\mathcal{R}\tilde{J}_{1,2,1}(\tilde{\omega}_3 + \alpha_{10})(\tilde{\lambda}_2 + \mathcal{R}) \\ &+ 2i\mathcal{R}\tilde{J}_{1,3,1}(\tilde{\omega}_3 + \alpha_{10}) + \tilde{L}_{1,1}(\mathcal{R}(\tilde{\lambda}_2 + \mathcal{R})(\alpha_{11}^2 \\ &+ i\mathcal{R}\tilde{\omega}_4) - 4i\alpha_{10}\alpha_{11}\mathcal{R}\tilde{\omega}_2 - 8\alpha_{10}^2\alpha_{11}^2)/2 \\ &+ \tilde{L}_{2,1}(\tilde{\lambda}_2(-2\alpha_{11}^2 - i\mathcal{R}\tilde{\omega}_4) + \mathcal{R}(-\alpha_{11}^2 - i\mathcal{R}\tilde{\omega}_4)) \\ &+ \mathcal{R}\tilde{\chi}_{1,1,3}(\tilde{\lambda}_2 + \mathcal{R})(2\alpha_{10}\alpha_{11} + i\mathcal{R}\tilde{\omega}_2)/2 \\ &+ \tilde{\chi}_{1,3,3}(8\alpha_{10}\alpha_{11} + 2i\mathcal{R}\tilde{\omega}_2) + \tilde{\chi}_{1,2,3}(-\mathcal{R}(2\alpha_{10}\alpha_{11} \\ &+ i\mathcal{R}\tilde{\omega}_2) - \tilde{\lambda}_2(4\alpha_{10}\alpha_{11} + i\mathcal{R}\tilde{\omega}_2)) + i\mathcal{R}\tilde{A}_1(\mathcal{R}\tilde{\lambda}_2\cdot \\ &\cdot(\tilde{\omega}_5 + \alpha_{11})/2 - \mathcal{R}^2\tilde{\omega}_5\alpha_{11}(2\alpha_{11}\tilde{\omega}_1 + 4\alpha_{10}(\tilde{\omega}_3 \\ &+ \alpha_{10}) - \mathcal{R}^2)) + i\mathcal{R}^2\tilde{\omega}_1\tilde{\phi}_{1,1,1}(\tilde{\lambda}_2 + \mathcal{R})/2 \\ &- i\mathcal{R}\tilde{\omega}_1\tilde{\phi}_{1,2,1}(\tilde{\lambda}_2 + \mathcal{R}) + 2i\mathcal{R}\tilde{\omega}_1\tilde{\phi}_{1,3,1},\end{aligned} \tag{C44}$$

$$\begin{aligned}\tilde{f}_{1,2,3} =& i\mathcal{R}\tilde{J}_{1,2,1}(\alpha_{10}(\mathcal{R}^2 - 4\alpha_{11}\tilde{\omega}_1) + \mathcal{R}\tilde{\lambda}_2(\tilde{\omega}_3 + \alpha_{10}) \\ &+ \mathcal{R}^2\tilde{\omega}_3)/2 + \tilde{L}_{2,1}(\mathcal{R}(\tilde{\lambda}_2 + \mathcal{R})(\alpha_{11}^2 + i\mathcal{R}\tilde{\omega}_4) \\ &- 4i\alpha_{10}\alpha_{11}\mathcal{R}\tilde{\omega}_2 - 8\alpha_{10}^2\alpha_{11}^2)/2 + \mathcal{R}\tilde{\chi}_{1,2,3}\cdot \\ &\cdot(\tilde{\lambda}_2 + \mathcal{R})(2\alpha_{10}\alpha_{11} + i\mathcal{R}\tilde{\omega}_2)/2 - 2i\mathcal{R}\tilde{J}_{1,3,1}\cdot \\ &\cdot(\tilde{\omega}_3 + \alpha_{10})(\tilde{\lambda}_2 + \mathcal{R}) + \tilde{\chi}_{1,3,3}(-2\tilde{\lambda}_2(4\alpha_{10}\alpha_{11} \\ &+ i\mathcal{R}\tilde{\omega}_2) - 2\mathcal{R}(2\alpha_{10}\alpha_{11} + i\mathcal{R}\tilde{\omega}_2)) + \tilde{\chi}_{1,4,3}\cdot \\ &\cdot(24\alpha_{10}\alpha_{11} + 6i\mathcal{R}\tilde{\omega}_2) + \frac{1}{2}i\mathcal{R}^2\tilde{\omega}_1\tilde{\phi}_{1,2,1}(\tilde{\lambda}_2 + \mathcal{R}) \\ &- 2i\mathcal{R}\tilde{\omega}_1\tilde{\phi}_{1,3,1}(\tilde{\lambda}_2 + \mathcal{R}) + 6i\mathcal{R}\tilde{\omega}_1\tilde{\phi}_{1,4,1},\end{aligned} \tag{C45}$$

$$\begin{aligned}\tilde{f}_{1,3,3} =& i\mathcal{R}\tilde{J}_{1,3,1}(\alpha_{10}(\mathcal{R}^2 - 4\alpha_{11}\tilde{\omega}_1) + \mathcal{R}\tilde{\lambda}_2(\tilde{\omega}_3 + \alpha_{10}) \\ &+ \mathcal{R}^2\tilde{\omega}_3)/2 + \mathcal{R}\tilde{\chi}_{1,3,3}(\tilde{\lambda}_2 + \mathcal{R})(2\alpha_{10}\alpha_{11} \\ &+ i\mathcal{R}\tilde{\omega}_2)/2 + \tilde{\chi}_{1,4,3}(-3\tilde{\lambda}_2(4\alpha_{10}\alpha_{11} + i\mathcal{R}\tilde{\omega}_2) \\ &- 3\mathcal{R}(2\alpha_{10}\alpha_{11} + i\mathcal{R}\tilde{\omega}_2)) + i\mathcal{R}^2\tilde{\omega}_1\tilde{\phi}_{1,3,1}(\tilde{\lambda}_2 \\ &+ \mathcal{R})/2 - 3i\mathcal{R}\tilde{\omega}_1\tilde{\phi}_{1,4,1}(\tilde{\lambda}_2 + \mathcal{R}) \\ &+ 12i\mathcal{R}\tilde{\omega}_1\tilde{\phi}_{1,5,1},\end{aligned} \tag{C46}$$

$$\begin{aligned}\tilde{f}_{1,4,3} =& \mathcal{R}(\tilde{\lambda}_2 + \mathcal{R})\Big(\tilde{\chi}_{1,4,3}(2\alpha_{10}\alpha_{11} + i\mathcal{R}\tilde{\omega}_2) \\ &+ i\mathcal{R}\tilde{\omega}_1\tilde{\phi}_{1,4,1} - 8i\tilde{\omega}_1\tilde{\phi}_{1,5,1}\Big)/2,\end{aligned} \tag{C47}$$

$$\tilde{f}_{1,5,3} = \frac{1}{2}i\mathcal{R}^2\tilde{\omega}_1\tilde{\phi}_{1,5,1}(\tilde{\lambda}_2 + \mathcal{R}), \tag{C48}$$

$$\begin{aligned}\tilde{f}_{1,1,4} =& -2i\alpha_{11}\alpha_{10}\mathcal{R}\tilde{\omega}_1\tilde{J}_{1,1,2} + 2i\mathcal{R}(\tilde{\omega}_3 + \alpha_{10})\cdot \\ &\cdot(\tilde{J}_{1,3,2} - \alpha_{10}\tilde{J}_{1,2,2}) - \alpha_{11}\alpha_{10}\tilde{L}_{1,2}(4\alpha_{10}\alpha_{11} \\ &+ \mathcal{R}(\alpha_{11} + 2i\tilde{\omega}_2)) - 2i\alpha_{10}\mathcal{R}\tilde{\omega}_1\tilde{\phi}_{1,2,3} + \tilde{L}_{2,2}\cdot \\ &\cdot(-2i\alpha_{10}\mathcal{R}\tilde{\omega}_4 + \alpha_{11}^2\mathcal{R} - 4\alpha_{10}\alpha_{11}^2) - 2\alpha_{11}\alpha_{10}^2\cdot \\ &\cdot\mathcal{R}\tilde{\chi}_{1,1,4} + \tilde{\chi}_{1,3,4}(8\alpha_{10}\alpha_{11} + 2i\mathcal{R}\tilde{\omega}_2) - i\alpha_{11}\mathcal{R}\cdot \\ &\cdot\tilde{A}_2(\alpha_{11}\tilde{\omega}_1 + 2\alpha_{10}(\tilde{\omega}_3 + \alpha_{10})) + 2\alpha_{10}\tilde{\chi}_{1,2,4}\cdot \\ &\cdot(\mathcal{R}(\alpha_{11} - i\tilde{\omega}_2) - 4\alpha_{10}\alpha_{11}) + 2i\mathcal{R}\tilde{\omega}_1\tilde{\phi}_{1,3,3},\end{aligned} \tag{C49}$$

$$\begin{aligned}\tilde{f}_{1,2,4} =& -2i\alpha_{11}\alpha_{10}\mathcal{R}\tilde{\omega}_1\tilde{J}_{1,2,2} - 4i\alpha_{10}\mathcal{R}\tilde{J}_{1,3,2}(\tilde{\omega}_3 \\ &+ \alpha_{10}) - \alpha_{10}\alpha_{11}\tilde{L}_{2,2}(4\alpha_{10}\alpha_{11} + \mathcal{R}(\alpha_{11} \\ &+ 2i\tilde{\omega}_2)) + 6i\mathcal{R}\tilde{\omega}_1\tilde{\phi}_{1,4,3}, + 4\alpha_{10}\tilde{\chi}_{1,3,4}(\mathcal{R}(\alpha_{11} \\ &- i\tilde{\omega}_2) - 4\alpha_{10}\alpha_{11}) - 2\alpha_{11}\alpha_{10}^2\mathcal{R}\tilde{\chi}_{1,2,4} + \tilde{\chi}_{1,4,4}\cdot \\ &\cdot(24\alpha_{10}\alpha_{11} + 6i\mathcal{R}\tilde{\omega}_2) - 4i\alpha_{10}\mathcal{R}\tilde{\omega}_1\tilde{\phi}_{1,3,3}\end{aligned} \tag{C50}$$

$$\begin{aligned}\tilde{f}_{1,3,4} =& -2i\alpha_{11}\alpha_{10}\mathcal{R}\tilde{\omega}_1\tilde{J}_{1,3,2} + 6\alpha_{10}\tilde{\chi}_{1,4,4}(-4\alpha_{10}\alpha_{11} \\ &+ \mathcal{R}(\alpha_{11} - i\tilde{\omega}_2)) - 6i\alpha_{10}\mathcal{R}\tilde{\omega}_1\tilde{\phi}_{1,4,3} \\ &- 2\alpha_{11}\alpha_{10}^2\mathcal{R}\tilde{\chi}_{1,3,4},\end{aligned} \tag{C51}$$

$$\tilde{f}_{1,4,4} = -2\alpha_{10}^2\alpha_{11}\mathcal{R}\tilde{\chi}_{1,4,4}, \tag{C52}$$

$$\begin{aligned}\tilde{f}_{2,1,1} =& -i\alpha_{10}\tilde{J}_{2,1,1} + \tilde{\chi}_{2,1,1}\left(2\alpha_{10}\alpha_{11} + i\tilde{\omega}_2\right) \\ &+ i\tilde{\omega}_1\tilde{\phi}_{2,1,2} - i\alpha_{11}\tilde{A}_3,\end{aligned} \tag{C53}$$

$$\tilde{f}_{2,2,1} = i\tilde{\omega}_1\tilde{\phi}_{2,2,2} - i\alpha_{10}\tilde{J}_{2,2,1}, \tag{C54}$$

$$\tilde{f}_{2,3,1} = -i\alpha_{10}\tilde{J}_{2,3,1}, \tag{C55}$$

$$\begin{aligned}\tilde{f}_{2,1,2} =& \delta\tilde{J}_{1,1,1} + \tilde{\chi}_{2,1,2}\left(2\alpha_{10}\alpha_{11} + i\tilde{\omega}_2\right) \\ &+ i\tilde{\omega}_1\tilde{\phi}_{2,1,3},\end{aligned} \tag{C56}$$

$$\tilde{f}_{2,2,2} = \delta\tilde{J}_{1,2,1} + i\tilde{\omega}_1\tilde{\phi}_{2,2,3}, \tag{C57}$$

$$\tilde{f}_{2,3,2} = \delta\tilde{J}_{1,3,1}, \tag{C58}$$

$$\begin{aligned}\tilde{f}_{2,1,3} =& \delta\tilde{J}_{1,1,2} + \tilde{\chi}_{2,1,3}\left(2\alpha_{10}\alpha_{11} + i\tilde{\omega}_2\right) \\ &+ i\tilde{\omega}_1\tilde{\phi}_{2,1,4},\end{aligned} \tag{C59}$$

$$\tilde{f}_{2,2,3} = \delta\tilde{J}_{1,2,2} + i\tilde{\omega}_1\tilde{\phi}_{2,2,4}, \tag{C60}$$

$$\tilde{f}_{2,3,3} = \delta\tilde{J}_{1,3,2}, \tag{C61}$$

$$\begin{aligned}\tilde{f}_{2,1,4} =& i\tilde{J}_{2,1,1}\left(\tilde{\omega}_3 + \alpha_{10}\right) + \tilde{P}_{1,1}\left(\alpha_{11}^2 + i\tilde{\omega}_4\right) \\ &+ \tilde{\chi}_{2,1,4}\left(2\alpha_{10}\alpha_{11} + i\tilde{\omega}_2\right) + i\tilde{\omega}_1\tilde{\phi}_{2,1,1} \\ &+ i\tilde{A}_3\left(\tilde{\omega}_5 + \alpha_{11}\right),\end{aligned} \tag{C62}$$

$$\begin{aligned}\tilde{f}_{2,2,4} =& i\tilde{J}_{2,2,1}\left(\tilde{\omega}_3 + \alpha_{10}\right) + \tilde{P}_{2,1}\left(\alpha_{11}^2 + i\tilde{\omega}_4\right) \\ &+ \tilde{\chi}_{2,2,4}\left(2\alpha_{10}\alpha_{11} + i\tilde{\omega}_2\right) + i\tilde{\omega}_1\tilde{\phi}_{2,2,1},\end{aligned} \tag{C63}$$

$$\begin{aligned}\tilde{f}_{2,3,4} =& i\tilde{J}_{2,3,1}\left(\tilde{\omega}_3 + \alpha_{10}\right) + \tilde{\chi}_{2,3,4}\left(2\alpha_{10}\alpha_{11} + i\tilde{\omega}_2\right) \\ &+ i\tilde{\omega}_1\tilde{\phi}_{2,3,1},\end{aligned} \tag{C64}$$

$$\tilde{f}_{2,4,4} = \tilde{\chi}_{2,4,4}\left(2\alpha_{10}\alpha_{11} + i\tilde{\omega}_2\right) + i\tilde{\omega}_1\tilde{\phi}_{2,4,1}, \tag{C65}$$

$$\tilde{f}_{2,5,4} = i\tilde{\omega}_1\tilde{\phi}_{2,5,1}. \tag{C66}$$

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