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Effect of surfactants on the inertialess instability of a two-layer film flow

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The effect of insoluble surface and interfacial surfactants on the inertialess instability of a two-fluid film flow down an inclined plane is investigated based on a normal mode analysis. The results reveal that the inertialess instability of relatively long waves can be predominantly weakened by a surface surfactant and enhanced by an interfacial surfactant. For sufficiently large viscosity ratio of the upper layer to the lower one, a destabilizing influence of the surface surfactant is also detected; this is thus a rare example demonstrating the possible destabilizing effect of the surfactant on the flow with a free surface. When the upper layer is less viscous and hence the instability due to the viscosity stratification disappears, a new instability can be triggered by the presence of an interfacial surfactant. Both the surfactants on the surface and the interface can stabilize or destabilize the short-wave instabilities, which occur for negligible surface and interfacial tensions.

1. Introduction

The gravity-driven flow of multiple-layer liquid films down an inclined plane is of considerable interest in fundamentals and applications. For example, a two-layer flow can be regarded as an elementary component in the coating processes encountered in the manufacturing of photographic films (Weinstein & Ruschak 2004). Instabilities of such a flow in the form of travelling surface and interfacial waves may lead to undesirable variations of the film thickness. In addition, most interfacial flows are accompanied by surface-active agents or surfactants, which usually play a critical role on flow stability. Thus, it is desirable to explore the stability of the multiple-layer flows and relevant influences of the surfactants.

The multiple-layer flows with a free surface can exhibit instability in the form of travelling waves. This type of instability does not need the support of fluid inertia and occurs even in the limit of Stokes flow; thus it is referred to as inertialess instability. This feature is different from the long-wave instability of a single-layer film, in which the inertia plays an important role in triggering the instability (Benjamin 1957; Yih 1963). Based on extensive studies, inertialess instabilities are encountered in two-layer flows (Kao 1968; Loewenherz & Lawrence 1989; Chen 1993) and a variety of flows with a configuration of more than two layers (Wang, Seaborg & Lin 1978; Weinstein & Kurz 1991; Weinstein & Chen 1999; Pozrikidis 2004). Nonlinear evolutions of the inertialess instability of multiple flows have been studied by Kliakhandler &

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Sivashinsky (1997) and Kliakhandler (1999) using a weakly nonlinear analysis and by Jiang *et al.* (2005) based on both numerical simulations and experiments.

Two-layer film flow has attracted much attention from workers owing to its simplicity and rich dynamics, even though film flows with multiple layers (more than two) are quite common in applications. Therefore, we focus mainly on two-layer film flow in the following. A linear stability of two-layer clean falling films with different viscosities and densities under a long-wave approximation was first investigated by Kao (1968). Two modes associated with the wave motions of the surface and interface are identified. The interface mode is unstable even at zero Reynolds number for the case of a more viscous upper layer. Loewenherz & Lawrence (1989) carried out a finite-wavelength stability analysis of the flow with matched densities and negligible surface/interfacial tension in the limit of Stokes flow, and focused on the role of viscosity stratification. They found that the flow is unstable and the most dangerous mode has a finite wavelength for the less viscous layer adjacent to the wall, whereas an inverse flow configuration is stable for all wavenumbers. Chen (1993) extended the stability analysis to include inertial effects and dealt with the effects of surface and interfacial tensions on the flow stability. Hu *et al.* (2006) performed an inertialess spatio-temporal stability analysis of the flow with combined effects of density and viscosity stratification. In addition, Jiang, Helenbrook & Lin (2004) revealed the important role of the interfacial shear work in the inertialess instability based on an energy budget approach.

The presence of an insoluble surfactant may have either a stabilizing or a destabilizing influence on the stability of interfacial flows. For a single-layer film flow with a free surface, the insoluble surfactants are stabilizing so that the critical Reynolds number increases (Whitaker & Jones 1966; Lin 1970; Blyth & Pozrikidis 2004*a*; Gao & Lu 2006). However, Wei (2005*a*, 2007) revealed that a single fluid film with an additional surface shear may also be destabilized by introducing an insoluble surfactant. For a two-fluid channel flow with a non-zero interfacial shear, an insoluble surfactant can induce an instability even in the limit of Stokes flow (Frenkel & Halpern 2002; Halpern & Frenkel 2003; Blyth & Pozrikidis 2004*b*). The underlying mechanism of the surfactant-induced instability was proposed by Wei (2005*b*) based on the viewpoint of vorticity. Destabilization of the core–annular flow was also studied by Wei, Halpern & Grotberg (2005).

Although the effect of insoluble surfactants on the flows with a single surface or interface has been widely studied and well understood, the effect of surfactants on multiple-layer films remains unclear. The purpose of the present work is to study the stability of the surfactant-laden two-fluid falling film, which consists of both a zero-shear surface and a non-zero-shear interface.

2. Flow configuration and the stability problem

Consider the gravity-driven film flow of two-fluid layers down an inclined plane which is tilted at an angle θ with respect to the horizontal direction (figure 1). The fluids are assumed to be incompressible and Newtonian. The upper layer is occupied by fluid 1 with mean thickness d_1 and dynamic viscosity μ_1 and is adjacent to the passive air, and the lower layer by fluid 2 with mean thickness d_2 and viscosity μ_2. For simplicity, the densities of both fluids are matched and denoted by ρ. Let $h_1(x^*, t^*)$ and $h_2(x^*, t^*)$ be the departures of the perturbed fluid–air surface and fluid–fluid interface from their mean locations, where t^* is time. Here and below we use an asterisk to denote dimensional quantities. Both the free surface and the interface are

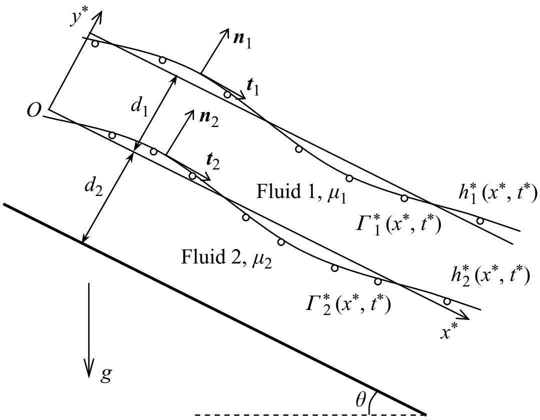

FIGURE 1. Schematic illustration of the two-layer flow down an inclined plane with surfactants.

covered by a monolayer of insoluble surfactants with concentrations $\Gamma_1^*(x^*, t^*)$ and $\Gamma_2^*(x^*, t^*)$, leading to variations of the surface tension $\gamma_1^*(x^*, t^*)$ and the interfacial tension $\gamma_2^*(x^*, t^*)$, respectively.

We assume the inertia of the fluid flow is negligible so that the governing equations for the motion of the flow are the Stokes equation and the continuity equation

$$-\nabla^* p_j^* + \mu_j \nabla^{*2} \boldsymbol{u}_j^* + \rho \boldsymbol{g} = 0, \qquad \nabla^* \cdot u_j^* = 0, \tag{2.1}$$

where $\nabla^* = (\partial/\partial x^*, \partial/\partial y^*)$, $\boldsymbol{u}_j^* = (u_j^*, v_j^*)$ is the fluid velocity, p_j^* is the pressure and $\boldsymbol{g}$ is the acceleration due to gravity. The subscripts $j = 1$ and 2 refer to the upper and lower fluid layer, respectively.

At the rigid wall ($y^* = -d_2$), the no-slip boundary condition, i.e. $\boldsymbol{u}^* = 0$, is employed. At the free surface ($y^* = d_1 + h_1^*$), the dynamic condition requiring the balance among the hydrodynamic traction, the surface tension and the Marangoni traction is

$$\boldsymbol{\sigma}_1^* \cdot \boldsymbol{n}_1 + (\gamma_1^* \nabla^* \cdot \boldsymbol{n}_1)\boldsymbol{n}_1 - \frac{1}{H_1}\frac{\partial \gamma_1^*}{\partial x^*}\boldsymbol{t}_1 = 0, \tag{2.2}$$

where $H_1 = \sqrt{1 + (\partial h_1^*/\partial x^*)^2}$, $\boldsymbol{\sigma}_1^*$ is the stress tensor, $\boldsymbol{n}_1$ is the unit normal vector pointing to the air, and $\boldsymbol{t}_1$ is the unit tangential vector pointing to the direction of increasing x^*. At the interface between the fluids ($y^* = h_2$), the velocity should be continuous, i.e. $\boldsymbol{u}_1^* = \boldsymbol{u}_2^*$, and the dynamic condition has the form

$$(\boldsymbol{\sigma}_1^* - \boldsymbol{\sigma}_2^*) \cdot \boldsymbol{n}_2 - (\gamma_2^* \nabla^* \cdot \boldsymbol{n}_2)\boldsymbol{n}_2 + \frac{1}{H_2}\frac{\partial \gamma_2^*}{\partial x^*}\boldsymbol{t}_2 = 0, \tag{2.3}$$

where $H_2 = \sqrt{1 + (\partial h_2^*/\partial x^*)^2}$, $\boldsymbol{n}_2$, pointing to fluid 1, and $\boldsymbol{t}_2$ are the unit vectors normal and tangential to the interface. In addition, we apply the kinematic condition

$$\frac{\partial h_j^*}{\partial t^*} + u_j^* \frac{\partial h_j^*}{\partial x^*} = v_j^* \tag{2.4}$$

at the free surface and the interface.

The concentrations of the insoluble surfactants, $\Gamma_j^*(x^*, t^*)$, are governed by a convection–diffusion equation (e.g. Halpern & Frenkel 2003), which for the one-dimensional case can be written as

$$\frac{\partial(H_j\Gamma_j^*)}{\partial t}+\frac{\partial}{\partial x^*}(H_j\Gamma_j^* u_j^*)=D_{sj}\frac{\partial}{\partial x^*}\left(\frac{1}{H_j}\frac{\partial \Gamma_j^*}{\partial x^*}\right), \tag{2.5}$$

where D_{sj} is the surfactant diffusivity. In practice, the surfactant diffusivity is typically negligible, so that we only account for the convection effect and take $D_{sj}=0$. Since we are concerned with infinitesimal perturbations for the linear stability problem, the relation between the surfactant concentrations Γ_j^* and the surface or interfacial tensions γ_j^* can be approximated as

$$\gamma_j^*-\gamma_{j0}=-E_j(\Gamma_j^*-\Gamma_{j0}), \tag{2.6}$$

where E_j is the surfactant elasticity, and Γ_{j0} is the basic value of the surfactant concentration, corresponding to the uniform surface or interfacial tension γ_{j0}.

Following Loewenherz & Lawrence (1989), we use the mean thickness of the lower layer, d_2, as the scale of length, and define the velocity scale as $\hat{U}=\rho g d_2^2\sin\theta/\mu_2$. The characteristic time and pressure are $d_2/\hat{U}$ and $\mu_2\hat{U}/d_2$, respectively. The surface and interfacial tensions as well as the surfactant concentrations are scaled by their basic values. In the basic state, the surface and the interface are flat, corresponding to uniform surfactant concentrations and surface tensions. The dimensionless velocity profile driven by gravity has the form

$$U_1(y)=m^{-1}\left(\delta y-\tfrac{1}{2}y^2\right)+\delta+\tfrac{1}{2}\quad\text{for}\quad 0\leqslant y\leqslant\delta, \tag{2.7}$$

$$U_2(y)=\delta y-\tfrac{1}{2}y^2+\delta+\tfrac{1}{2}\quad\text{for}\quad -1\leqslant y\leqslant 0, \tag{2.8}$$

where $m=\mu_1/\mu_2$ and $\delta=d_1/d_2$ are, respectively, the ratios of viscosities and thicknesses of the two layers. The basic pressure distributions are given by

$$P_1(y)=P_2(y)=(\delta-y)\cot\theta. \tag{2.9}$$

To study the linear stability of the basic state, the perturbed flow is decomposed into

$$(u_j,v_j)=(U_j,0)+(u_j',v_j'), \tag{2.10}$$

$$(p_j,\gamma_j,\Gamma_j)=(P_j,1,1)+(p_j',\gamma_j',\Gamma_j'). \tag{2.11}$$

Here a prime is used to denote perturbation qualities. Since only two-dimensional perturbations are considered, the continuity equations allow us to introduce the disturbance streamfunctions ψ_j', related to the velocity perturbations (u', v') as $u_j'=\partial\psi_j'/\partial y$, $v_j'=-\partial\psi_j'/\partial x$. Further, since the basic state is independent of x, the disturbances can be assumed to have the form of normal modes as

$$\begin{bmatrix}\psi_j'(x,y,t)\\ p_j'(x,y,t)\\ \gamma_j'(x,t)\\ \Gamma_j'(x,t)\\ h_j(x,t)\end{bmatrix}=\varepsilon\begin{bmatrix}\phi_j(y)\\ q_j(y)\\ \zeta_j\\ \xi_j\\ \eta_j\end{bmatrix}\mathrm{e}^{\mathrm{i}k(x-ct)}+\text{c.c.}, \tag{2.12}$$

where c.c. denotes the complex conjugate, $\varepsilon\ll 1$ is the infinitesimally small amplitude of the perturbations, k is a real streamwise wavenumber, and $c=c_r+\mathrm{i}c_i$ is the complex phase velocity of the disturbance wave.

Substituting the perturbations (2.12) into the dimensionless form of the governing equations (2.1), linearizing by retaining only the linear terms in ε, and eliminating the pressures, we have the Orr–Sommerfeld equations in the limit of Stokes flow

$$(\mathrm{D}^2 - k^2)^2\phi_j = 0, \tag{2.13}$$

for $j = 1, 2$, where $\mathrm{D} = \mathrm{d}/\mathrm{d}y$.

The no-slip boundary conditions at the rigid wall can be expressed in terms of ϕ as

$$\phi_2 = 0, \quad \mathrm{D}\phi_2 = 0 \quad \text{at} \quad y = -1. \tag{2.14}$$

The linearized versions of the boundary conditions at the free surface and the interface can be obtained by Taylor expanding the exact conditions in powers of h_j around their mean positions and then retaining the leading-order terms. At the fluid–fluid interface, the linearized boundary conditions associated with the continuity of the velocity and the dynamic conditions can be written as

$$\eta_2\mathrm{D}U_1 + \mathrm{D}\phi_1 = \eta_2\mathrm{D}U_2 + \mathrm{D}\phi_2, \tag{2.15}$$

$$\phi_1 = \phi_2, \tag{2.16}$$

$$m(\mathrm{D}^2 - 3k^2)\mathrm{D}\phi_1 = (\mathrm{D}^2 - 3k^2)\mathrm{D}\phi_2 - Ca_2^{-1}\mathrm{i}k^3\eta_2, \tag{2.17}$$

$$m(\mathrm{D}^2 + k^2)\phi_1 - (\mathrm{D}^2 + k^2)\phi_2 = Ma_2Ca_2^{-1}\mathrm{i}k\xi_2, \tag{2.18}$$

at $y = 0$. The dynamic conditions at the free surface become

$$\mathrm{i}k\eta_1\mathrm{D}P_1 + m(\mathrm{D}^2 - 3k^2)\mathrm{D}\phi_1 = Ca_1^{-1}\mathrm{i}k^3\eta_1, \tag{2.19}$$

$$\eta_1\mathrm{D}^2U_1 + (\mathrm{D}^2 + k^2)\phi_1 = -Ma_1m^{-1}Ca_1^{-1}\mathrm{i}k\xi_1, \tag{2.20}$$

at $y = \delta$. Finally, the linearized kinematic conditions and transport equations for the surfactants are

$$(U_1 - c)\eta_1 + \phi_1 = 0 \quad \text{at} \quad y = \delta, \tag{2.21}$$

$$(U_2 - c)\eta_2 + \phi_2 = 0 \quad \text{at} \quad y = 0, \tag{2.22}$$

$$(U_1 - c)\xi_1 + \mathrm{D}\phi_1 = 0 \quad \text{at} \quad y = \delta, \tag{2.23}$$

$$(U_2 - c)\xi_2 + \eta_2\mathrm{D}U_2 + \mathrm{D}\phi_2 = 0 \quad \text{at} \quad y = 0. \tag{2.24}$$

The additional four dimensionless numbers involved in the dynamic boundary conditions are defined as $Ma_j = E_j\Gamma_{j0}/\gamma_{j0}$ and $Ca_j = \mu_j\hat{U}/\gamma_{j0}$. Here, the parameters Ma_j are the Marangoni numbers, expressing the sensitivity of the surface and interfacial tensions on the surfactant concentrations, and Ca_j are the capillary numbers.

The general solutions to (2.13) have the form

$$\phi_j(y) = A_j\cosh ky + B_j\sinh ky + C_jy\cosh ky + D_jy\sinh ky, \tag{2.25}$$

where the coefficients A_j, B_j, C_j and D_j are to be determined. Upon substituting them into the boundary conditions (2.16) to (2.24), we obtain a homogeneous linear system, which can be written in the matrix form

$$\boldsymbol{M}\cdot w = 0, \tag{2.26}$$

where $\boldsymbol{w}=(A_1, B_1, C_1, D_1, A_2, B_2, C_2, D_2, \eta_1, \eta_2, \xi_1, \xi_2)^T$ is the unknown vector, and the coefficient matrix $\boldsymbol{M}$ is given by

$$\boldsymbol{M}=\begin{pmatrix}
0 & 0 & 0 & 0 & C_k & -S_k & -C_k & S_k & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & -kS_k & kC_k & C_k+kS_k & -kC_k-S_k & 0 & 0 & 0 & 0 \\
0 & k & 1 & 0 & 0 & -k & -1 & 0 & 0 & \dfrac{\delta}{m}-\delta & 0 & 0 \\
1 & 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & m & 0 & 0 & 0 & -1 & 0 & 0 & 0 & -\dfrac{\mathrm{i}k}{2Ca_2} & 0 & 0 \\
km & 0 & 0 & m & -k & 0 & 0 & -1 & 0 & 0 & 0 & -\dfrac{\mathrm{i}Ma_2}{2Ca_2} \\
2\mathrm{i}mk^2S_{k\delta} & 2\mathrm{i}mk^2C_{k\delta} & 2\mathrm{i}mk^2\delta S_{k\delta} & 2\mathrm{i}mk^2\delta C_{k\delta} & 0 & 0 & 0 & 0 & -\cot\theta-\dfrac{k^2}{Ca_1} & 0 & 0 & 0 \\
2k^2C_{k\delta} & 2k^2S_{k\delta} & 2k^2\delta C_{k\delta}+2kS_{k\delta} & 2kC_{k\delta}+2k^2\delta S_{k\delta} & 0 & 0 & 0 & 0 & -\dfrac{1}{m} & 0 & \dfrac{\mathrm{i}kMa_1}{mCa_1} & 0 \\
C_{k\delta} & S_{k\delta} & \delta C_{k\delta} & \delta S_{k\delta} & 0 & 0 & 0 & 0 & U_s-c & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & U_i-c & 0 & 0 \\
kS_{k\delta} & kC_{k\delta} & C_{k\delta}+k\delta S_{k\delta} & k\delta C_{k\delta}+S_{k\delta} & 0 & 0 & 0 & 0 & 0 & 0 & U_s-c & 0 \\
0 & 0 & 0 & 0 & 0 & k & 1 & 0 & 0 & \delta & 0 & U_i-c
\end{pmatrix},$$

where $C_k=\cosh k$, $S_k=\sinh k$, $C_{k\delta}=\cosh k\delta$, $S_{k\delta}=\sinh k\delta$, and U_s, U_i are the dimensionless basic velocities at the unperturbed surface and interface, respectively. A non-trivial solution of the system (2.26) exists if and only if the determinant of the 12×12 matrix $\boldsymbol{M}$ vanishes, which yields a dispersion relation between the wavenumber and the phase velocity for specified values of other parameters, given by

$$F(k, c; m, \delta, Ma_1, Ma_2, Ca_1, Ca_2)=0. \tag{2.27}$$

Since c appears only in the last four rows of $\boldsymbol{M}$, the dispersion relation (2.27) is a quartic equation for c, which is presented in the Appendix available as a supplement to the online version of this paper. Thus, we obtain four travelling-wave modes of the stability of the flow corresponding to the four roots of the dispersion relation. Two of the modes are associated with the deformation of the surface and the interface, which have been extensively investigated in previous studies (Kao 1968; Loewenherz & Lawrence 1989; Chen 1993; Hu *et al.* 2006), while the other two modes are related to the presence of the insoluble surfactants.

3. Results and discussion

In the absence of surfactants (i.e. $Ma_1 = Ma_2 = 0$), the last two rows and columns of $\boldsymbol{M}$ are irrelevant to the dynamics, and the flow admits only two normal modes. The growth rates of the unstable modes obtained are in excellent agreement with the known results (Loewenherz & Lawrence 1989; Chen 1993; Hu *et al.* 2006) for uncontaminated flows. Further, according to Blyth & Pozrikidis (2004*a*), the phase velocity of long-wave perturbations ($k \ll 1$) in the limit of Stokes flow for a single-layer falling film is given by

$$c = (\delta + 1)^2 - \mathrm{i}k \left[\tfrac{1}{3}(\delta + 1)^3 \cot\theta + \frac{Ma_1}{Ca_1}(\delta + 1) \right] + O(k^2). \tag{3.1}$$

To examine the reliability of the results in the presence of surfactant, calculations were performed for $m = 1$, $Ma_1 > 0$, $Ma_2 = 0$ and $Ca_2 = \infty$, corresponding to a surfactant-laden single-layer film flow; the values of c obtained by the present method agree well with the asymptotic predictions (3.1). To check the results for general parameters, the long-wave stability analysis has also been performed. By using a standard asymptotic procedure in the limit of $k \to 0$, as in Kao (1968), we obtain an eigenvalue problem for the long-wave instability

$$\boldsymbol{N} \cdot \boldsymbol{s} = c\boldsymbol{s}, \tag{3.2}$$

where $\boldsymbol{s} = (h_1, h_2, \xi_1, \xi_2)^T$ and

$$\boldsymbol{N} = \begin{pmatrix} 2U_s - \left(\dfrac{\delta^3}{3m} + \delta^2 + \delta + \tfrac{1}{3} \right) \mathrm{i}k \cot\theta & \delta^2 \left(1 - \dfrac{1}{m} \right) & -mU_s M_1 \mathrm{i}k & -U_i M_2 \mathrm{i}k \\ \tfrac{1}{2} - \left(\dfrac{\delta}{2} + \tfrac{1}{3} \right) \mathrm{i}k \cot\theta & U_i & -\tfrac{1}{2} m M_1 \mathrm{i}k & -\tfrac{1}{2} M_2 \mathrm{i}k \\ \left(\dfrac{\delta}{m} + 1 \right) - U_s \mathrm{i}k \cot\theta & \delta \left(1 - \dfrac{1}{m} \right) & U_s - (\delta + m) M_1 \mathrm{i}k & -M_2 \mathrm{i}k \\ 1 - U_i \mathrm{i}k \cot\theta & \delta & -m M_1 \mathrm{i}k & U_i - M_2 \mathrm{i}k \end{pmatrix},$$

with $M_1 = Ma_1/Ca_1$ and $M_2 = Ma_2/Ca_2$. Again, the results obtained by (2.27) for small k and arbitrary m are in excellent agreement with those predicted by the asymptotic procedure. By setting $m = 1$ and $M_2 = 0$, it can be proved that (3.1) is an eigenvalue of (3.2) as expected. However, since there are four degrees of freedom for general conditions instead of two as in the single-fluid surfactant problem, the phase velocity of long waves cannot be presented in forms as simple as (3.1). In addition, note that considering only the limit of long waves is insufficient to resolve the complete stability characteristics of the flow, which will be discussed below.

Based on our extensive calculations, at most one of the four modes may be unstable for a group of specified flow parameters. Here, we focus on the behaviour of the unstable modes and investigate their dependence on the surface and interfacial surfactants.

We first consider the case in which the free surface is covered by an insoluble surfactant while the interface remains clean. Figure 2 shows the variation of the growth rates kc_i of the unstable modes as a function of k for $\delta = 1$, $Ca_1 = Ca_2 = 1$, $Ma_2 = 0$, $\theta = 0.2$ and different values of Ma_1. For $m < 1$, no instability occurs and the relevant results are not shown. Here, the viscosity ratio is typically chosen as $m = 2.5$ (figure 2*a*) and $m = 50$ (figure 2*b*). Since the upper layer is more viscous than the lower layer, the flow is unstable in the absence of surfactant. For $m = 2.5$, it can be seen from figure 2(*a*) that the effect of surface surfactant is twofold. On the one hand, the

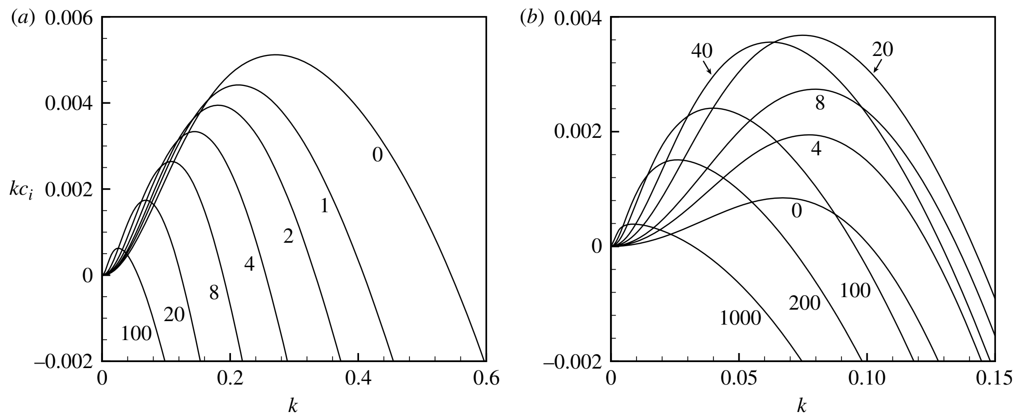

FIGURE 2. Effect of the surface surfactant on the stability for $\delta = 1$, $Ca_1 = Ca_2 = 1$, $Ma_2 = 0$, $\theta = 0.2$ and various values of Ma_1: (a) $m = 2.5$; (b) $m = 50$.

bandwidth of the unstable wavenumber is narrowed by the presence of surfactant; on the other hand, the maximum growth rate decreases monotonically with Ma_1 though the growth rates in very long waves are increased, and the corresponding wavelengths are shifted to the long-wave range. Hence the effect of the surface surfactant is stabilizing, consistent with previous findings in a single-fluid film flow (e.g. Blyth & Pozrikidis 2004a). As $Ma_1 \to \infty$, the maximum growth rate tends to zero owing to the surface immobilization. However, the inertialess instability cannot be fully eliminated though it is significantly weakened, since the sufficiently long waves are always unstable for all non-zero Ma_1.

For a much larger viscosity ratio $m = 50$, as shown in figure 2(b), the dependence of the maximum growth rate on Ma_1 exhibits a non-monotonic behaviour. As Ma_1 is raised from zero, the range of unstable wavenumbers is widened and the growth rates of the mode are significantly increased. Thus, the inertialess instability of the flow is enhanced by the presence of the surface surfactant. Beyond a threshold $Ma_1 \sim 20$, the growth rates begin to decrease and the unstable interval of k becomes smaller and smaller with increasing Ma_1. Because of the surface immobilization, the growth rates eventually tend to zero and hence the effect of surfactant is stabilizing for sufficiently large Ma_1 (e.g. $Ma_1 = 1000$). Note that the destabilizing influence of the surface surfactant on the stability of the flow at large viscosity ratio is a new finding, when considering the zero shear of the basic flow at the surface, since it is well known that surfactant plays an stabilizing role on the stability of a surfactant-laden free-surface flow. This behaviour should be because the unstable mode originates from the so-called interface mode at $Ma_1 = 0$, which is associated with the local dynamics near the interface, instead of the free surface. Hence, the introduction of an insoluble surface surfactant may be destabilizing.

Effects of the interfacial surfactant on the stability of the flow are shown in figure 3. The growth rate of the dominant mode for $m = 2.5$, $\delta = 1$, $Ca_1 = Ca_2 = 1$, $Ma_1 = 0$, $\theta = 0.2$ and various values of Ma_2 are shown in figure 3(a). The dependence of the growth rates on the interfacial surfactant is similar to that on the surface surfactant shown in figure 2(b). The main difference is that the variation of the growth rates is more sensitive to Ma_2, since the inertialess instability is associated with the presence of the interface, as discussed above. It is clear that the inertialess instability is enhanced by the interfacial surfactant for small and moderate values of Ma_2 and weakened for

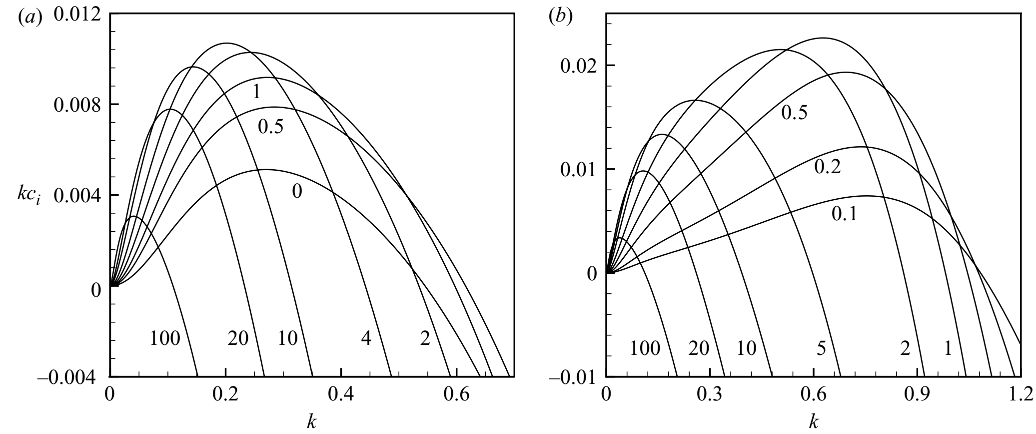

FIGURE 3. Effect of the interfacial surfactant on the stability for $\delta = 1$, $Ca_1 = Ca_2 = 1$, $Ma_1 = 0$, $\theta = 0.2$ and various values of Ma_2: (*a*) $m = 2.5$; (*b*) $m = 0.4$.

sufficiently large Ma_2 owing to the interface immobilization. The destabilizing effect of the interfacial surfactant is also relevant to the surfactant-induced instability of a two-layer channel flow found by Frenkel & Halpern (2002) and Halpern & Frenkel (2003), which occurs for a non-zero interfacial shear, as in the present case.

When a more viscous layer is adjacent to the plate, i.e. $m < 1$, the inertialess instability disappears if the interface is clean. By introducing an interfacial surfactant, an additional mode occurs and dominates the stability. The growth rates of the unstable mode are shown in figure 3(*b*) for $m = 0.4$ and various values of Ma_2 with other parameters the same as those in figure 3(*a*). It can be seen that this mode is unstable for all non-zero Ma_2 and the strongest instability occurs for $Ma_2 \sim O(1)$. As $Ma_2 \to 0$, the growth rate approaches zero and the cutoff wavenumber, beyond which the growth rate of the mode is negative, seems to tend to a finite value 1.08. It is emphasized that this type of instability is caused solely by the presence of the interfacial surfactant, since the effect of the viscosity stratification for $m < 1$ is always stabilizing.

Typical neutral stability curves in the (k, m)-plane are shown in figure 4(*a, b*) for $\delta = 1$, $Ca_1 = Ca_2 = 1$, $\theta = 0.2$ and different values of Ma_1 and Ma_2. In both the figures, the region to the left-hand side of each curve corresponds to unstable modes, and the region to the right-hand side side to stable modes, as denoted by U and S, respectively. Note that, when $Ma_1 = Ma_2 = 0$, the flow with equal viscosity is stable and the line $m = 1$ does not mean a neutral curve. In addition, the neutral curve doubles back at $k \approx 0.6$, so that all modes at larger wavenumbers are damped. These behaviours are due to the stabilizing effects of the surface and interfacial tensions – different from the results of Loewenherz & Lawrence (1989) and Hu *et al.* (2006) in which the tensions are completely neglected. For the flow with a surfactant-laden surface and a clean interface ($Ma_1 \neq 0$ and $Ma_2 = 0$) (figure 4*a*), all curves converge at the point $(k, m) = (0, 1)$. The flow for $m < 1$ is stable at all wavenumbers, the same as the case without surfactants. The curve has a single peak for small Ma_1 (e.g. $Ma_1 = 1$ and 2). As Ma_1 is increased to 8, the single peak splits into two peaks with one located near $m = 1$ and the other around $m = 10$. The cutoff wavenumber decreases monotonically with Ma_1 for typically $m < 10$, and increases for larger m. Remember that, as shown in figure 2(*b*), the inertialess instability is enhanced for sufficiently

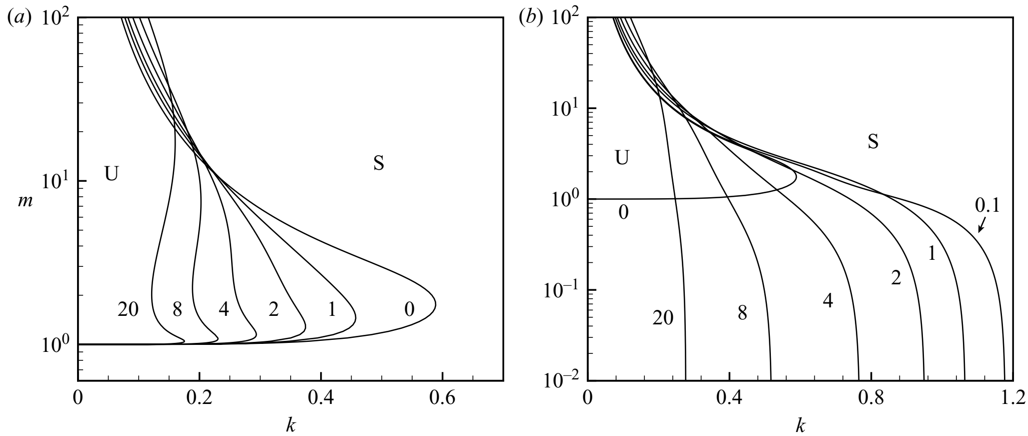

FIGURE 4. Neutral stability curves in the (k, m)-plane for (a) $Ma_2 = 0$ and different values of Ma_1, and (b) $Ma_1 = 0$ and different values of Ma_2. Other parameters are $\delta = 1$, $Ca_1 = Ca_2 = 1$ and $\theta = 0.2$. The stable and unstable regions are indicated by S and U, respectively.

large viscosity ratio. Neutral curves for the flow configuration with a free surface and surfactant-laden interface ($Ma_1 = 0$ and $Ma_2 \neq 0$) are shown in figure 4(b). The flow is unstable for all viscosity ratios. In particular, the instability persists even for $m = 1$, similar to the instability found by Frenkel & Halpern (2002) and Halpern & Frenkel (2003). The instability for $m < 1$ should be due primarily to the interaction between the surfactant and the interfacial shear, while, for $m > 1$, an additional mechanism of the inertialess instability in the absence of the surfactant is also responsible for the instability. Moreover, the cutoff wavenumber decreases monotonically with m once the interfacial surfactant is introduced.

We further investigate the effect of the surface surfactant on the instability induced by the interfacial surfactant for $m < 1$. The growth rates of the unstable mode for $m = 0.4$, $\delta = 1$, $Ca_1 = Ca_2 = 1$, $\theta = 0.2$ and different values of Ma_1 are shown in figure 5. We set Ma_2 to be unity so that a relatively strong instability can be obtained in the absence of the surface surfactant (see also figure 3b). As expected, the maximum growth rate is decreased with increasing Ma_1 and the instability is weakened. In addition, the curves for $Ma_1 > 100$ can barely be distinguished from the curve for $Ma_1 = 100$. This means that the maximum growth rate may tend to a finite value instead of zero as $Ma_1 \to \infty$ different from the results shown in figure 2, where the growth rate tends to zero. This behaviour is reasonably predicted since the interaction between the interfacial surfactant and the mean velocity shear is still active, even though the free surface becomes immobilized for sufficiently large Ma_1.

Finally, thanks to the comment and suggestion of a referee, it is of interest to study the limiting case of $Ca_1 = Ca_2 = \infty$ and finite Marangoni terms. The purpose is to isolate the Marangoni effects from the stabilizing influences of the surface and interfacial tensions. Accordingly, it is convenient to employ M_1 and M_2 as dimensionless parameters instead of Ma_1 and Ma_2, as in the long-wave analysis (3.2). The effect of the Marangoni traction on the inertialess instability is illustrated in figure 6, which shows the variations of the growth rates of the dominant mode as a function of k parameterized by M_1 and M_2. The capillary numbers are taken as $Ca_1 = Ca_2 = \infty$, indicating negligible surface and interfacial tension forces compared to the viscous forces and the Marangoni traction. In the absence of surfactants, the

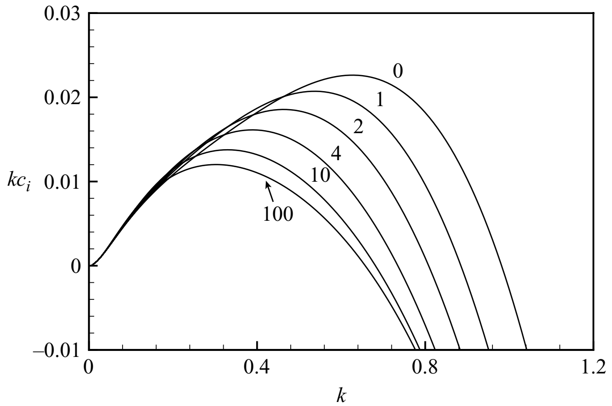

FIGURE 5. Damping effect of the surface surfactant on the instability induced by the interfacial surfactant for various values of Ma_1. Other parameters are $m = 0.4$, $\delta = 1$, $Ca_1 = Ca_2 = 1$, $Ma_2 = 1$ and $\theta = 0.2$.

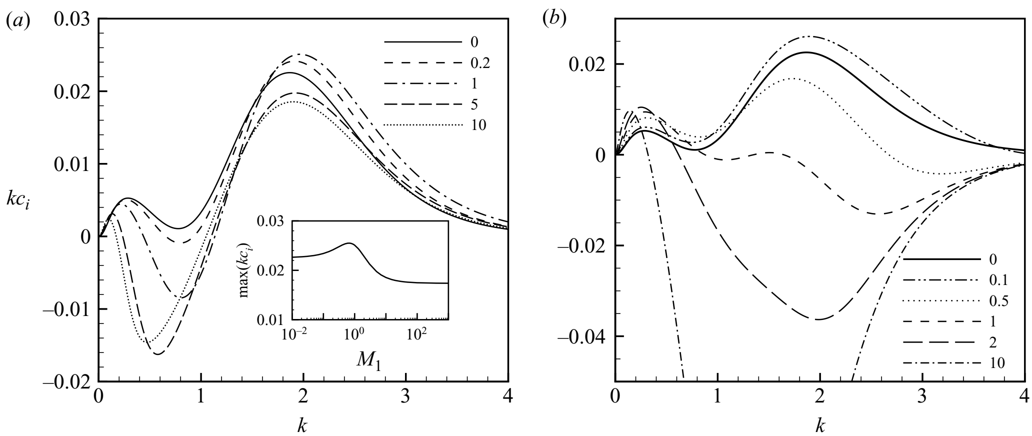

FIGURE 6. (*a*) Effects of surface surfactants on the inertialess instability for various values of M_1 and $M_2 = 0$; the maximum growth rates versus M_1 are plotted in the inset. (*b*) Effects of interfacial surfactants on the inertialess instability for $M_1 = 0$ and various values of M_2. The capillary numbers $Ca_1 = Ca_2 = \infty$ indicating negligible surface and interfacial tension forces. Other parameters are $m = 2.5$, $\delta = 1$ and $\theta = 0.2$.

growth rates exhibit two peaks, one of which corresponds to relatively long waves and the other to short waves, as also found by Loewenherz & Lawrence (1989). Note that the short-wave peak does not occur in the results discussed above owing to the stabilizing influences of the surface tensions. When the surface surfactant is introduced (figure 6*a*), the inertialess instability around the long-wave peaks is stabilized, similar to the results shown in figure 2(*a*). This behaviour is reasonable since the effects of surface tensions on long waves are always weak. Variation of the inertialess instability around the short-wave peaks indicates that the surface surfactant can be either stabilizing or destabilizing, as also revealed in the large-*m* case (see figure 2*b*). For clarity, the maximum growth rates as a function of M_1 are plotted in the inset of figure 6(*a*). The growth rates first increase with M_1 and then decreases when M_1 is larger than unity approximately. Note that the maximum growth rates tend to a finite value as $M_1 \to \infty$, indicating that the inertialess instability is present though the free surface is immobilized. Effects of the interfacial surfactants are demonstrated in figure 6(*b*). Again the modification of the long-wave instability is similar to that shown in figure 3(*a*) as expected. The variation of the short-wave peak is much more sensitive to M_2. For small values of M_2 (e.g. $M_2 = 0.1$), the peak moves upwards and hence the interfacial surfactants are destabilizing. As M_2 increases, the short-wave peak decreases rapidly and eventually disappears for large M_2 (e.g. $M_2 = 2$) together with the corresponding instability.

4. Conclusions

We have performed a linear stability analysis of a two-layer film flow laden by insoluble surface and interfacial surfactants in the limit of Stokes flow. Four normal modes associated with the stability are identified and only one exhibits instability. The results show that the growth of the unstable mode can be significantly affected by the surfactants. We first considered the case in which the surface and interfacial tensions are important so that short-wave instabilities are absent. For the viscosity ratio $m > 1$ but not much greater than unity, the effect of a surface surfactant tends to weaken the growth of the mode responsible for the inertialess instability and hence plays a stabilizing role on the flow. For $m \gg 1$, the growth rate of the unstable mode can be enhanced, indicating a destabilizing effect of the surface surfactant, in contrast to previous findings for surfactant-laden free-surface flow. The presence of an interfacial surfactant can promote the growth of the unstable mode for $m > 1$, and induces a new mode to dominate the instability for $m < 1$, leading to the occurrence of the instability for all viscosity ratios. When the surface surfactant is introduced, the instability caused by the interfacial surfactant for $m < 1$ is further weakened. For the case of negligible surface and interfacial tensions, short-wave instabilities can occur and dominate the flow. When M_1 and M_2 are small, the short-wave instabilities are enhanced; whereas it can be weakened by the surface surfactants and completely suppressed by the interfacial surfactants when M_1 and M_2 are large.

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REFERENCES

Benjamin, T. B. 1957 Wave formation in laminar flow down an inclined plane. *J. Fluid Mech.* **2**, 554–574.

Blyth, M. G. & Pozrikidis, C. 2004*a* Effect of surfactant on the stability of film flow down an inclined plane. *J. Fluid Mech.* **521**, 241–250.

Blyth, M. G. & Pozrikidis, C. 2004*b* Effect of surfactants on the stability of two-layer channel flow. *J. Fluid Mech.* **505**, 59–86.

Chen, K. P. 1993 Wave formation in the gravity-driven low-Reynolds number flow of two fluid films down an inclined plane. *Phys. Fluids* A **5**, 3038–3048.

Frenkel, A. L. & Halpern, D. 2002 Stokes-flow instability due to interfacial surfactant. *Phys. Fluids* **14**, L45–L48.

Gao, P. & Lu, X.-Y. 2006 Effect of surfactants on the long-wave stability of oscillatory film flow. *J. Fluid Mech.* **562**, 345–354.

Halpern, D. & Frenkel, A. L. 2003 Destabilization of a creeping flow by interfacial surfactant: linear theory extended to all wavenumbers. *J. Fluid Mech.* **485**, 191–220.

Hu, J., Millet, S., Botton, V., Hadid, H. B. & Henry, D. 2006 Inertialess temporal and spatio-temporal stability analysis of the two-layer film flow with density stratification. *Phys. Fluids* **18**, 104101.

Jiang, W. Y., Helenbrook, B. & Lin, S. P. 2004 Inertialess instability of a two-layer liquid film flow. *Phys. Fluids* **16**, 652–663.

Jiang, W. Y., Helenbrook, B. T., Lin, S. P. & Weinstein, S. J. 2005 Low-Reynolds-number instabilities in three-layer flow down an inclined wall. *J. Fluid Mech.* **539**, 387–416.

Kao, T. W. 1968 Role of viscosity stratification in the stability of two-layer flow down an incline. *J. Fluid Mech.* **33**, 561–572.

Kliakhandler, I. L. 1999 Long interfacial waves in multilayer thin films and coupled Kuramoto–Sivashinsky equations. *J. Fluid Mech.* **391**, 45–65.

Kliakhandler, I. L. & Sivashinsky, G. I. 1997 Viscous damping and instabilities in stratified liquid film flowing down a slightly inclined plane. *Phys. Fluids* **9**, 23–30.

Lin, S. P. 1970 Stabilizing effects of surface-active agents on a film flow. *AIChE J.* **16**, 375–379.

Loewenherz, D. S. & Lawrence, C. J. 1989 The effect of viscosity stratification on the stability of a free surface flow at low Reynolds number. *Phys. Fluids* A **1**, 1686–1693.

Pozrikidis, C. 2004 Instability of multi-layer channel and film flows. *Adv. Appl. Mech.* **40**, 179–239.

Wang, C. K., Seaborg, J. J. & Lin, S. P. 1978 Instability of multi-layered liquid films. *Phys. Fluids* **21**, 1669–1673.

Wei, H.-H. 2005*a* Effect of surfactant on the long-wave instability of a shear-imposed liquid flow down an inclined plane. *Phys. Fluids* **17**, 012103.

Wei, H.-H. 2005*b* On the flow-induced Marangoni instability due to the presence of surfactant. *J. Fluid Mech.* **544**, 173–200.

Wei, H.-H. 2007 Role of base flows on surfactant-driven interfacial instabilities. *Phys. Rev.* E **75**, 036306.

Wei, H.-H., Halpern, D. & Grotberg, J. B. 2005 Linear stability of a surfactant-laden annular film in a time-periodic pressure-driven flow through a capillary. *J. Colloid Interface Sci.* **285**, 769–780.

Weinstein, S. J. & Chen, K. P. 1999 Large growth rate instabilities in three-layer flow down an incline in the limit of zero Reynolds number. *Phys. Fluids* **11**, 3270–3282.

Weinstein, S. J. & Kurz, M. R. 1991 Long-wavelength instabilities in three-layer flow down an incline. *Phys. Fluids* A **3**, 2680–2687.

Weinstein, S. J. & Ruschak, K. J. 2004 Coating flows. *Annu. Rev. Fluid Mech.* **36**, 29–53.

Whitaker, S. & Jones, L. O. 1966 Stability of falling liquid films. Effect of interface and interfacial mass transport. *AIChE J.* **12**, 421–431.

Yih, C. S. 1963 Stability of liquid flow down an inclined plane. *Phys. Fluids* **6**, 321–334.

Appendix to "Effect of surfactants on the inertialess instability of a two-layer film flow"

By P. Gao & X.-Y. Lu

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Formulations of the quartic equation

The quartic equation for the phase velocity, i.e. c, can be given as

$$A_4c^4 + A_3c^3 + A_2c^2 + A_1c + A_0 = 0,$$

where A_i $(i = 0 \ldots 4)$ are the coefficients and can be expressed as follows:

$$A_0 = \left|\begin{array}{cccccccccccc}
0 & 0 & 0 & 0 & C_k & -S_k & -C_k & S_k & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & -kS_k & kC_k & C_k+kS_k & -kC_k-S_k & 0 & 0 & 0 & 0 \\
0 & k & 1 & 0 & 0 & -k & -1 & 0 & 0 & \frac{\delta}{m}-\delta & 0 & 0 \\
1 & 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & m & 0 & 0 & 0 & -1 & 0 & 0 & 0 & -\frac{\mathrm{i}k}{2Ca_2} & 0 & 0 \\
km & 0 & 0 & m & -k & 0 & 0 & -1 & 0 & 0 & 0 & -\frac{\mathrm{i}Ma_2}{2Ca_2} \\
2\mathrm{i}mk^2S_{k\delta} & 2\mathrm{i}mk^2C_{k\delta} & 2\mathrm{i}mk^2\delta S_{k\delta} & 2\mathrm{i}mk^2\delta C_{k\delta} & 0 & 0 & 0 & 0 & -\cot\theta-\frac{k^2}{Ca_1} & 0 & 0 & 0 \\
2k^2C_{k\delta} & 2k^2S_{k\delta} & 2k^2\delta C_{k\delta}+2kS_{k\delta} & 2kC_{k\delta}+2k^2\delta S_{k\delta} & 0 & 0 & 0 & 0 & -\frac{1}{m} & 0 & \frac{\mathrm{i}kMa_1}{mCa_1} & 0 \\
C_{k\delta} & S_{k\delta} & \delta C_{k\delta} & \delta S_{k\delta} & 0 & 0 & 0 & 0 & U_s & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & U_i & 0 & 0 \\
kS_{k\delta} & kC_{k\delta} & C_{k\delta}+k\delta S_{k\delta} & k\delta C_{k\delta}+S_{k\delta} & 0 & 0 & 0 & 0 & 0 & 0 & U_s & 0 \\
0 & 0 & 0 & 0 & 0 & k & 1 & 0 & 0 & \delta & 0 & U_i
\end{array}\right|;$$

$$A_1 = -\sum_{j=1}^{4} A_1^{(j)},$$

$$
A_1^{(1)} = \left| \begin{array}{ccccc}
0 & 0 & 0 & 0 & C_k \\
0 & 0 & 0 & 0 & -kS_k \\
0 & k & 1 & 0 & 0 \\
1 & 0 & 0 & 0 & -1 \\
0 & m & 0 & 0 & 0 \\
km & 0 & 0 & m & -k \\
2\mathrm{i}mk^2 S_{k\delta} & 2\mathrm{i}mk^2 C_{k\delta} & 2\mathrm{i}mk^2 \delta S_{k\delta} & 2\mathrm{i}mk^2 \delta C_{k\delta} & 0 \\
2k^2 C_{k\delta} & 2k^2 S_{k\delta} & 2k^2 \delta C_{k\delta} + 2kS_{k\delta} & 2kC_{k\delta} + 2k^2 \delta S_{k\delta} & 0 \\
0 & 0 & 0 & 0 & 1 \\
kS_{k\delta} & kC_{k\delta} & C_{k\delta} + k\delta S_{k\delta} & k\delta C_{k\delta} + S_{k\delta} & 0 \\
0 & 0 & 0 & 0 & 0
\end{array} \right.
$$

$$
\left. \begin{array}{cccccc}
-S_k & -C_k & S_k & 0 & 0 & 0 \\
kC_k & C_k + kS_k & -kC_k - S_k & 0 & 0 & 0 \\
-k & -1 & 0 & \frac{\delta}{m} - \delta & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 \\
-1 & 0 & 0 & -\frac{\mathrm{i}k}{2Ca_2} & 0 & 0 \\
0 & 0 & -1 & 0 & 0 & -\frac{\mathrm{i}Ma_2}{2Ca_2} \\
0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & \frac{\mathrm{i}kMa_1}{mCa_1} & 0 \\
0 & 0 & 0 & U_i & 0 & 0 \\
0 & 0 & 0 & 0 & U_s & 0 \\
k & 1 & 0 & \delta & 0 & U_i
\end{array} \right| ,
$$

$$
A_1^{(2)} = \left| \begin{array}{ccccc}
0 & 0 & 0 & 0 & C_k \\
0 & 0 & 0 & 0 & -kS_k \\
0 & k & 1 & 0 & 0 \\
1 & 0 & 0 & 0 & -1 \\
0 & m & 0 & 0 & 0 \\
km & 0 & 0 & m & -k \\
2\mathrm{i}mk^2 S_{k\delta} & 2\mathrm{i}mk^2 C_{k\delta} & 2\mathrm{i}mk^2 \delta S_{k\delta} & 2\mathrm{i}mk^2 \delta C_{k\delta} & 0 \\
2k^2 C_{k\delta} & 2k^2 S_{k\delta} & 2k^2 \delta C_{k\delta} + 2kS_{k\delta} & 2kC_{k\delta} + 2k^2 \delta S_{k\delta} & 0 \\
C_{k\delta} & S_{k\delta} & \delta C_{k\delta} & \delta S_{k\delta} & 0 \\
kS_{k\delta} & kC_{k\delta} & C_{k\delta} + k\delta S_{k\delta} & k\delta C_{k\delta} + S_{k\delta} & 0 \\
0 & 0 & 0 & 0 & 0
\end{array} \right.
$$

$$
\left. \begin{array}{cccccc}
-S_k & -C_k & S_k & 0 & 0 & 0 \\
kC_k & C_k + kS_k & -kC_k - S_k & 0 & 0 & 0 \\
-k & -1 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & 0 \\
-1 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & -1 & 0 & 0 & -\frac{\mathrm{i}Ma_2}{2Ca_2} \\
0 & 0 & 0 & -\cot\theta - \frac{k^2}{Ca_1} & 0 & 0 \\
0 & 0 & 0 & -\frac{1}{m} & \frac{\mathrm{i}kMa_1}{mCa_1} & 0 \\
0 & 0 & 0 & U_s & 0 & 0 \\
0 & 0 & 0 & 0 & U_s & 0 \\
k & 1 & 0 & 0 & 0 & U_i
\end{array} \right| ,
$$

$$A_1^{(3)} = \begin{vmatrix} 0 & 0 & 0 & 0 & C_k & -S_k & -C_k & S_k & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -kS_k & kC_k & C_k + kS_k & -kC_k - S_k & 0 & 0 & 0 \\ 0 & k & 1 & 0 & 0 & -k & -1 & 0 & 0 & \frac{\delta}{m} - \delta & 0 \\ 1 & 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & m & 0 & 0 & 0 & -1 & 0 & 0 & 0 & -\frac{\mathrm{i}k}{2Ca_2} & 0 \\ km & 0 & 0 & m & -k & 0 & 0 & -1 & 0 & 0 & -\frac{\mathrm{i}Ma_2}{2Ca_2} \\ 2\mathrm{i}mk^2 S_{k\delta} & 2\mathrm{i}mk^2 C_{k\delta} & 2\mathrm{i}mk^2 \delta S_{k\delta} & 2\mathrm{i}mk^2 \delta C_{k\delta} & 0 & 0 & 0 & 0 & -\cot\theta - \frac{k^2}{Ca_1} & 0 & 0 \\ 2k^2 C_{k\delta} & 2k^2 S_{k\delta} & 2k^2 \delta C_{k\delta} + 2kS_{k\delta} & 2kC_{k\delta} + 2k^2 \delta S_{k\delta} & 0 & 0 & 0 & 0 & -\frac{1}{m} & 0 & 0 \\ C_{k\delta} & S_{k\delta} & \delta C_{k\delta} & \delta S_{k\delta} & 0 & 0 & 0 & 0 & U_s & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & U_i & 0 \\ 0 & 0 & 0 & 0 & 0 & k & 1 & 0 & 0 & \delta & U_i \end{vmatrix},$$

$$A_1^{(4)} = \begin{vmatrix} 0 & 0 & 0 & 0 & C_k & -S_k & -C_k & S_k & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & -kS_k & kC_k & C_k + kS_k & -kC_k - S_k & 0 & 0 & 0 \\ 0 & k & 1 & 0 & 0 & -k & -1 & 0 & 0 & \frac{\delta}{m} - \delta & 0 \\ 1 & 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 & 0 & 0 \\ 0 & m & 0 & 0 & 0 & -1 & 0 & 0 & 0 & -\frac{\mathrm{i}k}{2Ca_2} & 0 \\ km & 0 & 0 & m & -k & 0 & 0 & -1 & 0 & 0 & 0 \\ 2\mathrm{i}mk^2 S_{k\delta} & 2\mathrm{i}mk^2 C_{k\delta} & 2\mathrm{i}mk^2 \delta S_{k\delta} & 2\mathrm{i}mk^2 \delta C_{k\delta} & 0 & 0 & 0 & 0 & -\cot\theta - \frac{k^2}{Ca_1} & 0 & 0 \\ 2k^2 C_{k\delta} & 2k^2 S_{k\delta} & 2k^2 \delta C_{k\delta} + 2kS_{k\delta} & 2kC_{k\delta} + 2k^2 \delta S_{k\delta} & 0 & 0 & 0 & 0 & -\frac{1}{m} & 0 & \frac{\mathrm{i}kMa_1}{mCa_1} \\ C_{k\delta} & S_{k\delta} & \delta C_{k\delta} & \delta S_{k\delta} & 0 & 0 & 0 & 0 & U_s & 0 & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & U_i & 0 \\ kS_{k\delta} & kC_{k\delta} & C_{k\delta} + k\delta S_{k\delta} & k\delta C_{k\delta} + S_{k\delta} & 0 & 0 & 0 & 0 & 0 & 0 & U_s \end{vmatrix};$$

$$A_2 = 2\sum_{j=1}^{6} A_2^{(j)},$$

$$A_2^{(1)} = \begin{vmatrix}
0 & 0 & 0 & 0 & C_k & -S_k & -C_k & S_k & 0 & 0 \\
0 & 0 & 0 & 0 & -kS_k & kC_k & C_k + kS_k & -kC_k - S_k & 0 & 0 \\
0 & k & 1 & 0 & 0 & -k & -1 & 0 & 0 & 0 \\
1 & 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 & 0 \\
0 & m & 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \\
km & 0 & 0 & m & -k & 0 & 0 & -1 & 0 & -\frac{\mathrm{i}Ma_2}{2Ca_2} \\
2\mathrm{i}mk^2 S_{k\delta} & 2\mathrm{i}mk^2 C_{k\delta} & 2\mathrm{i}mk^2\delta S_{k\delta} & 2\mathrm{i}mk^2\delta C_{k\delta} & 0 & 0 & 0 & 0 & 0 & 0 \\
2k^2 C_{k\delta} & 2k^2 S_{k\delta} & 2k^2\delta C_{k\delta} + 2kS_{k\delta} & 2kC_{k\delta} + 2k^2\delta S_{k\delta} & 0 & 0 & 0 & 0 & \frac{\mathrm{i}kMa_1}{mCa_1} & 0 \\
kS_{k\delta} & kC_{k\delta} & C_{k\delta} + k\delta S_{k\delta} & k\delta C_{k\delta} + S_{k\delta} & 0 & 0 & 0 & 0 & U_s & 0 \\
0 & 0 & 0 & 0 & 0 & k & 1 & 0 & 0 & U_i
\end{vmatrix},$$

$$A_2^{(2)} = \begin{vmatrix}
0 & 0 & 0 & 0 & C_k & -S_k & -C_k & S_k & 0 & 0 \\
0 & 0 & 0 & 0 & -kS_k & kC_k & C_k + kS_k & -kC_k - S_k & 0 & 0 \\
0 & k & 1 & 0 & 0 & -k & -1 & 0 & \frac{\delta}{m} - \delta & 0 \\
1 & 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 & 0 \\
0 & m & 0 & 0 & 0 & -1 & 0 & 0 & -\frac{\mathrm{i}k}{2Ca_2} & 0 \\
km & 0 & 0 & m & -k & 0 & 0 & -1 & 0 & -\frac{\mathrm{i}Ma_2}{2Ca_2} \\
2\mathrm{i}mk^2 S_{k\delta} & 2\mathrm{i}mk^2 C_{k\delta} & 2\mathrm{i}mk^2\delta S_{k\delta} & 2\mathrm{i}mk^2\delta C_{k\delta} & 0 & 0 & 0 & 0 & 0 & 0 \\
2k^2 C_{k\delta} & 2k^2 S_{k\delta} & 2k^2\delta C_{k\delta} + 2kS_{k\delta} & 2kC_{k\delta} + 2k^2\delta S_{k\delta} & 0 & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & U_i & 0 \\
0 & 0 & 0 & 0 & 0 & k & 1 & 0 & \delta & U_i
\end{vmatrix},$$

$$A_2^{(3)} = \left| \begin{matrix} 0 & 0 & 0 & 0 & C_k \\ 0 & 0 & 0 & 0 & -kS_k \\ 0 & k & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 & -1 \\ 0 & m & 0 & 0 & 0 \\ km & 0 & 0 & m & -k \\ 2\mathrm{i}mk^2 S_{k\delta} & 2\mathrm{i}mk^2 C_{k\delta} & 2\mathrm{i}mk^2\delta S_{k\delta} & 2\mathrm{i}mk^2\delta C_{k\delta} & 0 \\ 2k^2 C_{k\delta} & 2k^2 S_{k\delta} & 2k^2\delta C_{k\delta} + 2kS_{k\delta} & 2kC_{k\delta} + 2k^2\delta S_{k\delta} & 0 \\ 0 & 0 & 0 & 0 & 1 \\ kS_{k\delta} & kC_{k\delta} & C_{k\delta} + k\delta S_{k\delta} & k\delta C_{k\delta} + S_{k\delta} & 0 \end{matrix} \right.$$

$$\left. \begin{matrix} -S_k & -C_k & S_k & 0 & 0 \\ kC_k & C_k + kS_k & -kC_k - S_k & 0 & 0 \\ -k & -1 & 0 & \frac{\delta}{m} - \delta & 0 \\ 0 & 0 & 0 & 0 & 0 \\ -1 & 0 & 0 & -\frac{\mathrm{i}k}{2Ca_2} & 0 \\ 0 & 0 & -1 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & \frac{\mathrm{i}kMa_1}{mCa_1} \\ 0 & 0 & 0 & U_i & 0 \\ 0 & 0 & 0 & 0 & U_s \end{matrix} \right| ,$$

$$A_2^{(4)} = \left| \begin{matrix} 0 & 0 & 0 & 0 & C_k \\ 0 & 0 & 0 & 0 & -kS_k \\ 0 & k & 1 & 0 & 0 \\ 1 & 0 & 0 & 0 & -1 \\ 0 & m & 0 & 0 & 0 \\ km & 0 & 0 & m & -k \\ 2\mathrm{i}mk^2 S_{k\delta} & 2\mathrm{i}mk^2 C_{k\delta} & 2\mathrm{i}mk^2\delta S_{k\delta} & 2\mathrm{i}mk^2\delta C_{k\delta} & 0 \\ 2k^2 C_{k\delta} & 2k^2 S_{k\delta} & 2k^2\delta C_{k\delta} + 2kS_{k\delta} & 2kC_{k\delta} + 2k^2\delta S_{k\delta} & 0 \\ C_{k\delta} & S_{k\delta} & \delta C_{k\delta} & \delta S_{k\delta} & 0 \\ 0 & 0 & 0 & 0 & 0 \end{matrix} \right.$$

$$\left. \begin{matrix} -S_k & -C_k & S_k & 0 & 0 \\ kC_k & C_k + kS_k & -kC_k - S_k & 0 & 0 \\ -k & -1 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ -1 & 0 & 0 & 0 & 0 \\ 0 & 0 & -1 & 0 & -\frac{\mathrm{i}Ma_2}{2Ca_2} \\ 0 & 0 & 0 & -\cot\theta - \frac{k^2}{Ca_1} & 0 \\ 0 & 0 & 0 & -\frac{1}{m} & 0 \\ 0 & 0 & 0 & U_s & 0 \\ k & 1 & 0 & 0 & U_i \end{matrix} \right| ,$$

$$A_2^{(5)} = \begin{vmatrix} 0 & 0 & 0 & 0 & C_k & -S_k & -C_k & S_k & 0 & 0 \\ 0 & 0 & 0 & 0 & -kS_k & kC_k & C_k + kS_k & -kC_k - S_k & 0 & 0 \\ 0 & k & 1 & 0 & 0 & -k & -1 & 0 & 0 & 0 \\ 1 & 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 & 0 \\ 0 & m & 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \\ km & 0 & 0 & m & -k & 0 & 0 & -1 & 0 & 0 \\ 2\mathrm{i}mk^2 S_{k\delta} & 2\mathrm{i}mk^2 C_{k\delta} & 2\mathrm{i}mk^2 \delta S_{k\delta} & 2\mathrm{i}mk^2 \delta C_{k\delta} & 0 & 0 & 0 & 0 & -\cot\theta - \frac{k^2}{Ca_1} & 0 \\ 2k^2 C_{k\delta} & 2k^2 S_{k\delta} & 2k^2 \delta C_{k\delta} + 2kS_{k\delta} & 2kC_{k\delta} + 2k^2 \delta S_{k\delta} & 0 & 0 & 0 & 0 & -\frac{1}{m} & \frac{\mathrm{i}kMa_1}{mCa_1} \\ C_{k\delta} & S_{k\delta} & \delta C_{k\delta} & \delta S_{k\delta} & 0 & 0 & 0 & 0 & U_s & 0 \\ kS_{k\delta} & kC_{k\delta} & C_{k\delta} + k\delta S_{k\delta} & k\delta C_{k\delta} + S_{k\delta} & 0 & 0 & 0 & 0 & 0 & U_s \end{vmatrix},$$

$$A_2^{(6)} = \begin{vmatrix} 0 & 0 & 0 & 0 & C_k & -S_k & -C_k & S_k & 0 & 0 \\ 0 & 0 & 0 & 0 & -kS_k & kC_k & C_k + kS_k & -kC_k - S_k & 0 & 0 \\ 0 & k & 1 & 0 & 0 & -k & -1 & 0 & 0 & \frac{\delta}{m} - \delta \\ 1 & 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 & 0 \\ 0 & m & 0 & 0 & 0 & -1 & 0 & 0 & 0 & -\frac{\mathrm{i}k}{2Ca_2} \\ km & 0 & 0 & m & -k & 0 & 0 & -1 & 0 & 0 \\ 2\mathrm{i}mk^2 S_{k\delta} & 2\mathrm{i}mk^2 C_{k\delta} & 2\mathrm{i}mk^2 \delta S_{k\delta} & 2\mathrm{i}mk^2 \delta C_{k\delta} & 0 & 0 & 0 & 0 & -\cot\theta - \frac{k^2}{Ca_1} & 0 \\ 2k^2 C_{k\delta} & 2k^2 S_{k\delta} & 2k^2 \delta C_{k\delta} + 2kS_{k\delta} & 2kC_{k\delta} + 2k^2 \delta S_{k\delta} & 0 & 0 & 0 & 0 & -\frac{1}{m} & 0 \\ C_{k\delta} & S_{k\delta} & \delta C_{k\delta} & \delta S_{k\delta} & 0 & 0 & 0 & 0 & U_s & 0 \\ 0 & 0 & 0 & 0 & 1 & 0 & 0 & 0 & 0 & U_i \end{vmatrix};$$

$$A_3 = -\sum_{j=1}^{4} A_3^{(j)},$$

$$A_3^{(1)} = \begin{vmatrix}
0 & 0 & 0 & 0 & C_k & -S_k & -C_k & S_k & 0 \\
0 & 0 & 0 & 0 & -kS_k & kC_k & C_k + kS_k & -kC_k - S_k & 0 \\
0 & k & 1 & 0 & 0 & -k & -1 & 0 & 0 \\
1 & 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \\
0 & m & 0 & 0 & 0 & -1 & 0 & 0 & 0 \\
km & 0 & 0 & m & -k & 0 & 0 & -1 & -\frac{\mathrm{i}Ma_2}{2Ca_2} \\
2\mathrm{i}mk^2S_{k\delta} & 2\mathrm{i}mk^2C_{k\delta} & 2\mathrm{i}mk^2\delta S_{k\delta} & 2\mathrm{i}mk^2\delta C_{k\delta} & 0 & 0 & 0 & 0 & 0 \\
2k^2C_{k\delta} & 2k^2S_{k\delta} & 2k^2\delta C_{k\delta} + 2kS_{k\delta} & 2kC_{k\delta} + 2k^2\delta S_{k\delta} & 0 & 0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 & 0 & k & 1 & 0 & U_i
\end{vmatrix},$$

$$A_3^{(2)} = \begin{vmatrix}
0 & 0 & 0 & 0 & C_k & -S_k & -C_k & S_k & 0 \\
0 & 0 & 0 & 0 & -kS_k & kC_k & C_k + kS_k & -kC_k - S_k & 0 \\
0 & k & 1 & 0 & 0 & -k & -1 & 0 & 0 \\
1 & 0 & 0 & 0 & -1 & 0 & 0 & 0 & 0 \\
0 & m & 0 & 0 & 0 & -1 & 0 & 0 & 0 \\
km & 0 & 0 & m & -k & 0 & 0 & -1 & 0 \\
2\mathrm{i}mk^2S_{k\delta} & 2\mathrm{i}mk^2C_{k\delta} & 2\mathrm{i}mk^2\delta S_{k\delta} & 2\mathrm{i}mk^2\delta C_{k\delta} & 0 & 0 & 0 & 0 & 0 \\
2k^2C_{k\delta} & 2k^2S_{k\delta} & 2k^2\delta C_{k\delta} + 2kS_{k\delta} & 2kC_{k\delta} + 2k^2\delta S_{k\delta} & 0 & 0 & 0 & 0 & \frac{\mathrm{i}kMa_1}{mCa_1} \\
kS_{k\delta} & kC_{k\delta} & C_{k\delta} + k\delta S_{k\delta} & k\delta C_{k\delta} + S_{k\delta} & 0 & 0 & 0 & 0 & U_s
\end{vmatrix},$$

$$A_3^{(3)} = \left| \begin{matrix}
0 & 0 & 0 & 0 & C_k \\
0 & 0 & 0 & 0 & -kS_k \\
0 & k & 1 & 0 & 0 \\
1 & 0 & 0 & 0 & -1 \\
0 & m & 0 & 0 & 0 \\
km & 0 & 0 & m & -k \\
2\mathrm{i}mk^2S_{k\delta} & 2\mathrm{i}mk^2C_{k\delta} & 2\mathrm{i}mk^2\delta S_{k\delta} & 2\mathrm{i}mk^2\delta C_{k\delta} & 0 \\
2k^2C_{k\delta} & 2k^2S_{k\delta} & 2k^2\delta C_{k\delta} + 2kS_{k\delta} & 2kC_{k\delta} + 2k^2\delta S_{k\delta} & 0 \\
0 & 0 & 0 & 0 & 1
\end{matrix} \right.$$

$$
\left.\begin{array}{cccc}
-S_k & -C_k & S_k & 0 \\
kC_k & C_k + kS_k & -kC_k - S_k & 0 \\
-k & -1 & 0 & \frac{\delta}{m} - \delta \\
0 & 0 & 0 & 0 \\
-1 & 0 & 0 & -\frac{\mathrm{i}k}{2Ca_2} \\
0 & 0 & -1 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & U_i
\end{array}\right|,
$$

$$
A_3^{(4)} = \left|\begin{array}{ccccc}
0 & 0 & 0 & 0 & C_k \\
0 & 0 & 0 & 0 & -kS_k \\
0 & k & 1 & 0 & 0 \\
1 & 0 & 0 & 0 & -1 \\
0 & m & 0 & 0 & 0 \\
km & 0 & 0 & m & -k \\
2\mathrm{i}mk^2 S_{k\delta} & 2\mathrm{i}mk^2 C_{k\delta} & 2\mathrm{i}mk^2 \delta S_{k\delta} & 2\mathrm{i}mk^2 \delta C_{k\delta} & 0 \\
2k^2 C_{k\delta} & 2k^2 S_{k\delta} & 2k^2 \delta C_{k\delta} + 2kS_{k\delta} & 2kC_{k\delta} + 2k^2 \delta S_{k\delta} & 0 \\
C_{k\delta} & S_{k\delta} & \delta C_{k\delta} & \delta S_{k\delta} & 0
\end{array}\right.
$$

$$
\left.\begin{array}{cccc}
-S_k & -C_k & S_k & 0 \\
kC_k & C_k + kS_k & -kC_k - S_k & 0 \\
-k & -1 & 0 & 0 \\
0 & 0 & 0 & 0 \\
-1 & 0 & 0 & 0 \\
0 & 0 & -1 & 0 \\
0 & 0 & 0 & -\cot\theta - \frac{k^2}{Ca_1} \\
0 & 0 & 0 & -\frac{1}{m} \\
0 & 0 & 0 & U_s
\end{array}\right|;
$$

and

$$
A_4 = \left|\begin{array}{cccc}
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0 \\
0 & k & 1 & 0 \\
1 & 0 & 0 & 0 \\
0 & m & 0 & 0 \\
km & 0 & 0 & m \\
2\mathrm{i}mk^2 S_{k\delta} & 2\mathrm{i}mk^2 C_{k\delta} & 2\mathrm{i}mk^2 \delta S_{k\delta} & 2\mathrm{i}mk^2 \delta C_{k\delta} \\
2k^2 C_{k\delta} & 2k^2 S_{k\delta} & 2k^2 \delta C_{k\delta} + 2kS_{k\delta} & 2kC_{k\delta} + 2k^2 \delta S_{k\delta}
\end{array}\right.
$$

$$
\left.\begin{array}{cccc}
C_k & -S_k & -C_k & S_k \\
-kS_k & kC_k & C_k + kS_k & -kC_k - S_k \\
0 & -k & -1 & 0 \\
-1 & 0 & 0 & 0 \\
0 & -1 & 0 & 0 \\
-k & 0 & 0 & -1 \\
0 & 0 & 0 & 0 \\
0 & 0 & 0 & 0
\end{array}\right|.
$$