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Thermodynamic and kinetic instabilities of lattice-matched alloy layers: Compositional and morphological perturbations

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We study the compositional and morphological instabilities affecting epitaxial alloy layers lattice-matched to their substrate and alloy half spaces under zero net strain. We first calculate analytically the elastically relaxed state of such a system where a composition modulation is coupled to a surface undulation having either the same wavelength or half the wavelength of the composition modulation. The calculations, carried out to second order in undulation amplitude, prove that the coupling energy is nonzero in both cases. Analytical results for undulations of finite amplitude are also obtained. We then use these calculations to study in a unified fashion the thermodynamic and kinetic instabilities of these systems and to determine how compositional and morphological perturbations affect each other. We find that, at variance with the case of lattice-mismatched layers, the critical temperature for thermodynamic compositional instability of nongrowing layers is the same for planar and nonplanar surfaces. For growing layers, we show that the kinetic compositional instability may develop independently of any morphological perturbation. However, whether it is growing or not, a lattice-matched alloy with a finite lateral composition modulation is always unstable against some undulations of its free surface.

I. INTRODUCTION

Composition variations and free-surface nonplanarities are two perturbations that commonly affect epitaxial alloy layers. It is of both fundamental and practical interest to know if such layers are unstable with respect to such perturbations. In this context, two types of instability are now often considered. The notion of *thermodynamic* instability applies to a *static* (nongrowing) layer, a closed system containing a fixed number of atoms of each species, which is deemed unstable when the total energy of some perturbed state is less than the energy of a reference state, in which the alloy is homogeneous and has a planar free surface. This has to be distinguished from *kinetic* instabilities, which will only develop in the open system constituted by a *growing* layer onto which new material is continuously added.[1] We adopt these terms in the following although thermodynamic instabilities can obviously only develop via kinetic processes and kinetic instabilities have thermodynamic causes. As far as composition is concerned, thermodynamic instabilities can actually develop only if bulk atomic diffusion is effective, whereas kinetic instabilities require only surface diffusion. On the other hand, the development of morphological instabilities does not require bulk diffusion, so that for them the distinction between thermodynamic and kinetic instabilities is less pertinent.

Since in many alloys composition variations induce stresses, careful consideration of the elastic effects is necessary, whatever the type of instability considered. After compositional and morphological perturbations had been studied independently for many years, it was demonstrated recently that, in the case of a lattice-mismatched layer, they modify each other. In particular, the thermodynamic instability of such a layer with respect to a composition modulation (CM) coupled to a surface undulation (SU) differs greatly from its instability with respect to each perturbation considered independently.[2] The reason for this coupling is that in this case both perturbations induce elastic stress fields which interact. In the present work, we concentrate on layers that are lattice matched to their substrate in their unperturbed homogeneous state. Elastic coupling is then not obvious at first, since the morphological perturbation alone does not generate stresses. We show that it still exists in this case, however, and results in an extension of the domain of morphological instability, whereas it does not affect the proper kinetic compositional instability.

In Sec. II, we recall briefly the simple thermodynamical description used for the alloy and situate our study in the context of experimental results and previous theoretical approaches. In Sec. III, we calculate analytically to order 2 in SU amplitude the coherent stress relaxation and the energy for an alloy layer where the SU has either the wavelength or half the wavelength of the CM. In Sec. IV, we discuss the thermodynamic stability of such a layer against pure and coupled compositional and morphological perturbations. We also show that in some cases exact results can be obtained for surface perturbations of arbitrary amplitudes. Finally, we show in Sec. V that, although they do not take growth processes explicitly into account, our calculations provide a very simple demonstration of the pure compositional kinetic instability, besides allowing the inclusion of surface corrugations in the discussion of the latter.

II. PRESENTATION OF THE PROBLEM

A. Thermodynamic description of the alloy

Although the effects studied are not expected to depend on the particular description chosen for the alloy, their dis-

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cussion is simplified by specifying one. We summarize here the basic hypotheses retained. We consider a binary A_cB_{1-c} or pseudobinary $A_cB_{1-c}C$ cubic alloy whose stress-free (SF) lattice parameter a depends on composition c according to Vegard's law with $a^{-1}da/dc=\eta\neq 0$. The bulk and the surface of the alloy are treated as continua. With applications to semiconductors in mind, we suppose that the volume density of the chemical and entropic contributions to the free enthalpy is given by the regular solution model:[3]

$$\omega(c)=\Omega c(1-c)+RT[c\ln c+(1-c)\ln(1-c)], \quad (1)$$

where c is the local alloy composition, T the temperature, R the gas constant, and Ω the interaction parameter. The energy of a portion S of free surface is ΓS, with Γ the surface energy. η, Ω, and Γ are supposed independent of composition, orientation, and stress. Any composition inhomogeneity induces a spatial variation of the SF strain. In this work, we consider the coherent relaxation of the latter, and in particular the nonuniform relaxation near the free surface, using isotropic elasticity with Young modulus E and Poisson ratio ν.

B. Independent morphological and compositional instabilities and joint instabilities in lattice-mismatched layers

The first theoretical studies of alloys with a free surface were devoted to thermodynamic instabilities affecting independently either composition or surface morphology. When surface nonplanarities are ignored, the alloy of average composition $\overline{c}$ may be unstable with respect to CM's under some critical temperature if $\Omega>0$. We originally evaluated the critical temperature relative to CM's with wave vectors parallel to the surface in an elastically isotropic epitaxial layer (mismatched or not).[3] Ipatova, Malyshkin, and Shchukin[4] extended the calculation to arbitrary CM's in a semi-infinite elastically anisotropic half space. A similar calculation in the isotropic case yields

$$T_c^i(\overline{c})=(1-E\eta^2/2\Omega)T_c^b(\overline{c}), \quad (2)$$

where $T_c^b(\overline{c})=2\overline{c}(1-\overline{c})\Omega/R$ is the "chemical" critical temperature for the bulk alloy without elastic effects ($\eta=0$). These critical temperatures are maximum for $c=0.5$. When on the other hand the possible composition variations are ignored, one may apply the general result[5,6] for a half space submitted to biaxial stress $\overline{\sigma}$ or for an epitaxial layer lattice mismatched by $\overline{\epsilon}$ with respect to its substrate, which thus induces a biaxial stress $\overline{\sigma}=-E\overline{\epsilon}/(1-\nu)$: the planar free surface is then unstable with respect to SU's with wave numbers less than a critical value $k_c=2(1+\nu)(1-\nu)E^{-1}\Gamma^{-1}\overline{\sigma}^2=2(1+\nu)(1-\nu)^{-1}E\Gamma^{-1}\overline{\epsilon}^2$.

The proportionality of $T_c^b-T_c^i$ and k_c to E signals that elastic effects play a prominent part in both compositional and morphological instabilities. Indeed, they induce the latter whereas they inhibit the former. It is only recently, however, that the possible coupling of the two instabilities has been investigated. In a misfitting alloy layer with joint CM and SU, coupling arises because the elastic energy is not simply the sum of those associated separately with each perturbation. This question was investigated from the point of view of kinetics by Guyer and Voorhees[7] and Léonard and Desai,[8] who studied in particular the effects of the deposition rate and sign of the misfit on the homogeneity and planarity of the growing layer. We adopted a different approach:[2] after calculating the energy of a misfitting compositionally modulated and nonplanar film [Fig. 1(a)], we determined in which parameter range the homogeneous planar system is thermodynamically unstable with respect to coupled CM's and SU's having the same wave number, irrespective of the particular growth and diffusion conditions that may or may not allow it actually to become inhomogeneous and nonplanar. Whereas when coupling is ignored, the system is separately unstable with respect to CM's of arbitrary wave number for $T<T_c^i$ and to SU's for $k<k_c$ at any temperature, when coupling is duly taken into account, it is unstable at any temperature to a range $0<k<\tilde{k}_c(T)$ of joint CM's and SU's having the same wave number k.

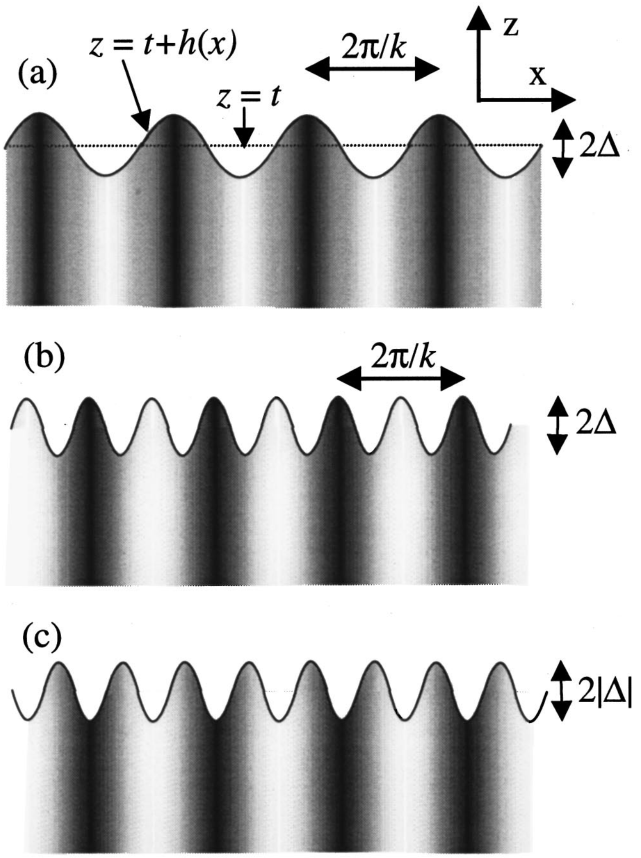

FIG. 1. Schematics of composition modulation coupled to surface undulation with (a) identical wavelength, and with half wavelength with (b) crests and (c) troughs at composition extrema (black and white).

C. Coupling of composition modulation and surface undulation in lattice-matched layers: Introduction

In mismatched layers, SU's are frequently observed[9] and the coupling between a CM and a SU with the same wave number has been reported.[10] This is interpretable in the theoretical framework recalled above. On the other hand, long-wavelength CM's have repeatedly been observed in apparently flat lattice-matched layers and it has been demonstrated that they appear during growth well above $T_c^i(c_0)$,c_0 being the composition of the lattice-matched film.[11] Moreover, correlated CM's and SU's have recently been reported in nominally lattice-matched films.[12,13] From the thermodynamic point of view, these observations are puzzling because, according to the previous models, CM's are then energetically unfavorable both for planar surfaces, for which T_c^i is relevant, and for undulated ones, since $\tilde{k}_c(T)$ vanishes with $\bar{\epsilon}$.

Several theoretical attempts have been made to explain the observation of CM's for $\bar{\epsilon}=0$. A first kinetic approach neglects possible surface nonplanarities and considers the development of a CM in material deposited on an already modulated underlying layer (the existence of which can be somewhat justified by the statistical alloy composition fluctuations). This follows our original suggestion[3] that, once a CM is initiated, it remains frozen under the newly deposited material but might be amplified in the latter due to the preferential attachment of like atoms to like atoms on the surface. Malyshkin and Shchukin[1] first calculated the corresponding kinetic instability temperature, higher than T_c^i and even than T_c^b. Ipatova *et al.*[14] recently developed this idea in detailed calculations. The dependence of the latter results on several poorly known quantities makes the comparison with experiments difficult.

A second type of approach considers the coupling of a CM with a SU, the underlying assumption being that, in samples where CM's have been observed, a small-amplitude SU might have remained undetected. Guyer and Voorhees[15] predicted a kinetic joint instability below a ''deposition'' critical temperature, which we found to coincide, somewhat surprisingly, with the critical temperature for pure compositional instability.[1] The results of Ref. 15 were obtained under the hypothesis of local vapor/solid equilibrium, the validity of which remains to be assessed for the various growth methods. Léonard and Desai[8] also studied the problem taking growth and diffusion into account but without resorting to local equilibrium. For nongrowing lattice-matched films, they find the planar surface stable at any temperature and the alloy compositionally unstable only below $T_c^i(\bar{c})$ [the introduction of a critical value of η in Ref. 8 is simply another way of expressing our Eq. (2)]. They also predict the planar surface of growing films to be always stable so that, for these authors, there is no specifically kinetic joint instability for lattice-matched alloy films. Finally, Venezuela and Tersoff[16] concluded that a possible kinetic coupled instability exists when the atoms involved have different surface mobilities, without, however, considering the compositional stresses (which is equivalent to setting $\eta=0$).

In view of these diverging results and unexpected coincidences, we reexamine here the compositional and morphological thermodynamic and kinetic instabilities of lattice-matched epitaxial layers and alloy half spaces under zero net stress (a thick plastically relaxed alloy layer grown on a lattice-mismatched substrate is a close approximation of the latter). In such studies, it is essential to calculate the elastically relaxed state of the system. For undulated surfaces, this can generally be done only to a given order in SU amplitude. Up to now, all calculations of the elastic fields in alloys have been performed to order 1 in SU amplitude (calculations to order 2 exist for homogeneous materials[17]) and for CM's and SU's having the same wave vector. This is adequate for $\bar{\epsilon}\neq 0$ since the first-order stress then couples to the average strain,[2] whereas for $\bar{\epsilon}=0$ the elastic energy of the system contains no coupling term. However, we expect some elastic coupling to subsist even then since the relaxation of the compositionally modulated thin film is partly determined by the boundary conditions on the free surface and cannot be the same whether the latter is planar or not (if $\eta\neq 0$). We thus consider a lattice-matched system with joint CM and SU, having identical or different wavelengths, and first calculate the first terms nonlinear in SU amplitude of the strain and stress fields and the total energy of the system, before showing that these results provide a means of studying in a unified fashion the questions of stability.

III. CALCULATION OF THE ELASTICALLY RELAXED STATE AND OF THE ENERGY OF THE PERTURBATION

A. Composition modulation and surface undulation with the same wavelength

We first study the coupling of a CM with a SU having the same wave number k [Fig. 1(a)]. This is by far the case most frequently considered theoretically and encountered experimentally. Consider a CM $c(x,z)=c_0+\eta^{-1}\epsilon_c m(z)\sin kx$ inducing a SF strain modulation

$$\epsilon_{ij}^{\mathrm{SF}}(x,z)=\delta_{ij}\epsilon_c m(z)\sin kx. \tag{3}$$

Here, m describes the depth dependence of the CM amplitude.[2] This formulation encompasses alloy half spaces but also alloy layers grown on a substrate extending up to $z=t_0$ [$m(z)=0$ for $z\leqslant t_0$], although distinction is unnecessary in practice for layer thicknesses large with respect to the wavelengths of the perturbations considered.[2] The surface undulates around average height t: $z=t+h(x)=t+\Delta\sin kx$. We first calculate the coherent stress relaxation with respect to the SF state when CM (3) occurs in a half space with planar free surface at $z=t+\Delta$:[2,18]

$$\begin{aligned}
\sigma_{xx}^{p}={}&-2F(4m(z)-2k\{[2k(z-t-\Delta)+3]I(k,t+\Delta)\\
&+I(k,z)+G(k,z,t+\Delta)\}e^{kz})\epsilon_c\sin kx,\\
\sigma_{yy}^{p}={}&-2F[4m(z)-8\nu kI(k,t+\Delta)e^{kz}]\epsilon_c\sin kx,\\
\sigma_{zz}^{p}={}&-4Fk\{[2k(z-t-\Delta)-1]I(k,t+\Delta)+I(k,z)\\
&+G(k,z,t+\Delta)\}\epsilon_c e^{kz}\sin kx,\\
\sigma_{xz}^{p}={}&-4Fk\{[2k(z-t-\Delta)+1]I(k,t+\Delta)-I(k,z)\\
&+G(k,z,t+\Delta)\}\epsilon_c e^{kz}\cos kx,
\end{aligned} \tag{4}$$

with $F=E/8(1-\nu)$ and

$$I(k,z)=e^{-kz}\int_{-\infty}^{z}m(\xi)e^{k(\xi-z)}d\xi,$$

$$G(k,z_1,z_2)=\int_{z_1}^{z_2}m(\xi)e^{-k\xi}d\xi. \tag{5}$$

These formulas are valid for any CM amplitude ϵ_c provided linear elasticity applies. In the presence of the SU, the boundary conditions become $\sigma_{ij}n_j=0$ on the undulated surface for the total stress field. Since σ_{ij}^{p} usually does not satisfy these conditions, additional stresses $\sigma_{ij}^{A}=\sigma_{ij}-\sigma_{ij}^{p}$ appear, which can be taken as deriving from an Airy stress function

$$\chi(x,z)=\sum_{n}[(A_n+B_n z)\sin nkx + (A_n'+B_n'z)\cos nkx]e^{nk(z-t)}:$$

$$\sigma_{xx}^{A}=k\sum_{n} n[(nkA_n+2B_n+nkB_n z)\sin nkx + (nkA_n'+2B_n'+nkB_n'z)\cos nkx]e^{nk(z-t)},$$

$$\sigma_{xz}^{A}=k\sum_{n} n[-(nkA_n+B_n+nkB_n z)\cos nkx + (nkA_n'+B_n'+nkB_n'z)\sin nkx]e^{nk(z-t)},$$

$$\sigma_{zz}^{A}=-k^2\sum_{n} n^2[(A_n+B_n z)\sin nkx + (A_n'+B_n'z)\cos nkx]e^{nk(z-t)}. \tag{6}$$

The calculations can usually only be performed at a given order in Δ. Since $\sigma_{ij}^{A}=0$ for $\Delta=0$, the coefficients A_n, B_n, A_n', B_n' are of order 1 at least. Since the elastic energy must be even in Δ, we perform calculations to order 2 (Appendix A) which show that in order to satisfy the boundary conditions terms up to $n=3$ must be included in χ. The only coefficients of order 1 or 2 are

$$A_1=A_3/2=FP(t)\epsilon_c\Delta^2,$$

$$B_1=2F\{-4P(t)+[3P(t)-Q(t)]k\Delta\}\epsilon_c\Delta,$$

$$B_3=2F[2P(t)-Q(t)]\epsilon_c k\Delta^2, \quad B_2'=-4FP(t)\epsilon_c\Delta, \tag{7}$$

with $P(t)=m(t)-2kI(k,t)e^{kt}$ and $2Q(t)=k^{-1}m'(t)-m(t)$. Equations (6), (7), and (4) [valid for any Δ and developed as Eq. (A2)] solve the problem at order 2 in Δ.

The elastic energy per unit surface is[3,2]

$$W_e=\frac{k}{4\pi}\int_0^{2\pi/k}\int_{-\infty}^{h(x)} -\epsilon_{ij}^{\mathrm{SF}}(x,z)(\sigma_{ij}^{p}+\sigma_{ij}^{A})(x,z)\,dx\,dz. \tag{8}$$

To second order in Δ,

$$-\epsilon_{ij}^{\mathrm{SF}}(\sigma_{ij}^{p}+\sigma_{ij}^{A})=2F\epsilon_c^2 m(z)\Big(4m(z)(1-\cos 2kx) + (1+\nu)\{-4J_1(t)+[P(t) - 3Q(t)]k^2\Delta^2\}e^{k(z-t)}(1-\cos 2kx) + 4(1+\nu)e^{2k(z-t)}P(t)k\Delta(\sin 3kx - \sin kx) - 3(1+\nu)e^{3k(z-t)}k^2\Delta^2[2P(t) - Q(t)](\cos 2kx-\cos 4kx)\Big)$$

and

$$W_e=4F\epsilon_c^2 k^{-1}[J_2(t)-(1+\nu)J_1^2(t)]+F\epsilon_c^2 k\Delta^2\{(1+\nu)J_1(t) \times[5m(t)-3k^{-1}m'(t)-2J_1(t)]-2(1+\nu)m^2(t) + 3m(t)k^{-1}m'(t)\} \tag{9}$$

with

$$J_n(t)=k\int_{-\infty}^{t} m^n(z)e^{(2-n)k(z-t)}dz \quad \text{for } n=1,2. \tag{10}$$

From Eq. (1), the chemical energy comprises a volume density $\omega(c_0)$ and a surface density

$$W_c=\frac{k}{2\pi}\int_0^{2\pi/k}\int_{-\infty}^{t+h(x)}\left\{\left[\Omega(1-2c_0)+RT\ln\left(\frac{c_0}{1-c_0}\right)\right]\delta c + \left(-\Omega+\frac{RT}{2c_0(1-c_0)}\right)(\delta c)^2\right\}dx\,dz, \tag{11}$$

where $\delta c(x,z)=c(x,z)-c_0$. To second order in Δ,

$$\int_0^{2\pi/k}\int_{-\infty}^{t+h(x)}\delta c\,dx\,dz=\pi k^{-1}\eta^{-1}m(t)\epsilon_c\Delta,$$

$$\int_0^{2\pi/k}\int_{-\infty}^{t+h(x)}(\delta c)^2 dx\,dz=\pi k^{-2}\eta^{-2} \times[J_2+\tfrac{3}{4}m(t)m'(t)k\Delta^2]\epsilon_c^2.$$

In the homogeneous planar reference state, the chemical energy comprises volume density $\omega(c_0)$ and surface density $W_c^{\mathrm{ref}}=\{\Omega(1-2c_0)+RT\ln[c_0/(1-c_0)]\}m(t)\epsilon_c\Delta/2\eta$ canceling exactly the first-order term of W_c, whereas the elastic energy is zero. The difference of free enthalpy per unit surface between the state with joint CM and SU and this reference state is thus

$$\delta H_1(h,\epsilon_c,\Delta)=\tfrac{1}{2}k^{-1}[\alpha(t)+\gamma(t)k^2\Delta^2]\epsilon_c^2+\tfrac{1}{4}\Gamma k^2\Delta^2 \tag{12}$$

with

$$\alpha(t)=4(1-\nu)F\left(J_2(t)\frac{\delta T}{\delta T_0}+\frac{1+\nu}{1-\nu}[J_2(t)-2J_1^2(t)]\right),$$

$$\gamma(t)=(1-\nu)F\left(3m(t)k^{-1}m'(t)\frac{\delta T}{\delta T_0} + \frac{1+\nu}{1-\nu}P(t)e^{-kt}[6Q(t)-P(t)e^{-kt}]\right),$$

$$\delta T=T-T_c^i(c_0). \quad \delta T_0=T_c^b(c_0)-T_c^i(c_0). \tag{13}$$

Equation (12) is exact in ϵ_c as long as linear elasticity applies. This should hold in practice because the CM's measured in coherent epitaxial layers induce fairly weak SF strains (below 1%).

B. Surface undulation with half the wavelength of the composition modulation

The coupling of a CM with a SU having half its wavelength [Figs. 1(b) and 1(c)] is also worth investigating. Considering independently the chemical and elastic energies suggests that such coupling might destabilize the alloy more than the coupling studied in Sec. III A. From the purely chemical point of view ($\eta=0$), a compositionally modulated alloy is trivially unstable for $T<T_c^i(c_0)=T_c^b(c_0)$ with respect to SU's having crests in the regions of high and low deviation from c_0, simply because such SU's increase the volume of material with nonaverage composition in the then compositionally unstable alloy. The reverse holds for $T>T_c^i(c_0)$. The kinetic aspects of this problem were investigated by Léonard and Desai.[19] Conversely, if $\eta\neq 0$, when a CM appears in a planar half space under zero net stress, the regions of maximal deviation from the average composition are the most highly stressed (under tension *and* compression). A SU with crests in these regions and troughs in the nearly lattice-matched zones should be favored with respect to a SU having troughs in the high-stress regions, since crests are known from the study of the standard morphological instability to be regions of easy elastic relaxation.

Consider [Figs. 1(b) and 1(c)] a SU $z=t+h(x)=t+\Delta\sin 2kx$ coupled to a CM inducing a SF strain modulation with extrema at the crests: $\epsilon_{ij}^{\mathrm{SF}}(x,z)=\delta_{ij}m(z)(\sin kx+\cos kx)$. $\Delta>0$ and $\Delta<0$ correspond, respectively, to maxima [Fig. 1(b)] and minima [Fig. 1(c)] at the crests. The calculation proceeds as in Sec. III A. Stresses in the planar half space $z\leqslant t+\Delta$ are obtained by replacing $\sin kx$ and $\cos kx$ by $(\sin kx+\cos kx)$ and $(\cos kx-\sin kx)$ in Eq. (4). From the calculations at order 2 in Δ made in Appendix B,

$$A_1=A_1'=2A_5=2A_5'=-2A_3=2A_3'=2FP(t)\epsilon_c\Delta^2,$$

$$B_1=B_1'=-2F\{2P(t)+[3P(t)+2Q(t)]k\Delta\}\epsilon_c\Delta,$$

$$B_3=-B_3'=2F\{2P(t)+[P(t)/6+Q(t)]k\Delta\}\epsilon_c\Delta,$$

$$B_5=B_5'=2F[31P(t)/10-Q(t)]\epsilon_c k\Delta^2. \tag{14}$$

The elastic energy is given by Eq. (8). To second order in Δ,

$$\begin{aligned}-\epsilon_{ij}^{\mathrm{SF}}(\sigma_{ij}^{p}+\sigma_{ij}^{A})={}&16F\epsilon_c^2m^2(z)(1+\sin 2kz)+4(1+\nu)F\epsilon_c^2\{-4J_1(t)-2P(t)k\Delta+[7P(t)-2Q(t)]k^2\Delta^2\}m(z)e^{k(z-t)}\\&\times(1+\sin 2kx)-2(1+\nu)F\epsilon_c^2\{12P(t)+k\Delta[P(t)+6Q(t)]\}k\Delta m(z)e^{3k(z-t)}(\sin 2kx-\cos 4kx)\\&-2(1+\nu)F\epsilon_c^2k^2\Delta^2[31P(t)-10Q(t)]m(z)e^{5k(z-t)}(\cos 4kx+\sin 6kx)\end{aligned}$$

and

$$\begin{aligned}W_e={}&8F\epsilon_c^2k^{-1}[J_2(t)-(1+\nu)J_1^2(t)]+4F\epsilon_c^2\Delta[m^2(t)\\&-2(1+\nu)m(t)J_1(t)+2(1+\nu)J_1^2(t)]\\&+2F\epsilon_c^2k\Delta^2\{2m(t)k^{-1}m'(t)-4(1+\nu)m^2(t)\\&+(1+\nu)J_1(t)[15m(t)-2k^{-1}m'(t)+14J_1(t)]\}.\end{aligned}\tag{15}$$

The surface density of chemical energy is given by Eq. (11). To second order in Δ.

$$\int_0^{2\pi/k}\int_{-\infty}^{t+h(x)}\delta c\,dx\,dz=0,$$

$$\begin{aligned}\int_0^{2\pi/k}\int_{-\infty}^{t+h(x)}(\delta c)^2dx\,dz={}&\pi k^{-2}\eta^{-2}[2J_2+m^2(t)k\Delta\\&+m(t)k^{-1}m'(t)k^2\Delta^2]\epsilon_c^2.\end{aligned}$$

Since the CM wavelength is twice the SU wavelength, the composition of the reference state is *exactly* c_0 so that the surface densities of elastic energy and chemical energy vanish. Hence, the free-enthalpy difference between the perturbed state and the reference state is

$$\begin{aligned}\delta H_2(t,\epsilon_c,\Delta)={}&k^{-1}[\alpha'(t)+\beta'(t)k\Delta+\gamma'(t)k^2\Delta^2]\epsilon_c^2\\&+\Gamma k^2\Delta^2\end{aligned}\tag{16}$$

with

$$\alpha'(t)=\alpha(t),$$

$$\beta'(t)=2(1-\nu)F\left(m^2(t)\frac{\delta T}{\delta T_0}+\frac{1+\nu}{1-\nu}P^2(t)\right),$$

$$\begin{aligned}\gamma'(t)=2(1-\nu)F\Bigg(&m(t)k^{-1}m'(t)\frac{\delta T}{\delta T_0}\\&+\frac{1+\nu}{1-\nu}\{m(t)k^{-1}m'(t)-2m^2(t)+J_1(t)[15m(t)\\&-2k^{-1}m'(t)+14J_1(t)]\}\Bigg).\end{aligned}\tag{17}$$

IV. THERMODYNAMIC COMPOSITIONAL INSTABILITY AND MORPHOLOGICAL INSTABILITY

We first consider the implications of the results of Sec. III for the thermodynamic stability of static films, for which the relevant energy is δH_1 or δH_2 (since stability is gauged with respect to the reference state) and for morphological instability. It should be borne in mind that in inhomogeneous films a SU always alters the composition distribution so that it is usually impossible to distinguish the energetic effect of the SU proper from the effect of this alteration.

A. Composition modulation and surface undulation with the same wavelength

Equation (12) shows that CM and SU are coupled. The coupling coefficient is $\gamma=\gamma_{\mathrm{ch}}+\gamma_e$, with

$\gamma_{\rm ch}=3(1-\nu)Fm(t)k^{-1}m'(t)\delta T/\delta T_0$ and $\gamma_e=(1+\nu)FP(t)e^{-kt}\lfloor 6Q(t)-P(t)e^{-kt}\rfloor$. $\gamma_{\rm ch}$ describes the effect of the purely compositional change induced by the SU and γ_e its elastic effects. As expected, the coupling energy is of order 2 in Δ and in ϵ_c. Consequently, the stability with respect to CM and SU (joint or not) of the homogeneous planar alloy layer is governed by α. This coefficient is the same as that found in the analysis of the planar half space with CM only.[2,4] The Schwarz theorem proves that $J_1^2(t) \leqslant J_2(t)/2$ for any function m, equality being reached for CM $m(z)=e^{kz}$, which we shall call critical. Hence, for $T > T_c^i(c_0)$, the homogeneous alloy is thermodynamically stable with respect to both CM and SU.

For $T<T_c^i(c_0)$, the system is thermodynamically unstable with respect to all CM's such that $\alpha<0$ (including the critical CM). At a given temperature, this is a condition on the depth dependence m of the CM. The range of unstable CM's increases when T decreases. The result is that a system where a CM of finite amplitude ϵ_c has developed is in addition unstable with respect to a SU, with the same wavelength provided $\gamma<0$ and $2k^{-1}\Gamma^{-1}|\gamma|\epsilon_c^2>1$. For T given, this is a condition on the amplitude, the wavelength, and the depth dependence of the CM. Such modulations exist: considering, for instance, the class of CM's $m(z)=(1+rz)e^{kz}$, one readily finds a range of values of r for which $\alpha<0$ and $\gamma<0$. The condition $2k^{-1}\Gamma^{-1}|\gamma|\epsilon_c^2>1$ links this result to the usual morphological instability[5,6] and to the coupled compositional instability of misfitting alloy layers,[2] since it can be rewritten as $k<\hat{k}_c(T)$, where $\hat{k}_c(T)$ is a temperature-dependent critical wave vector. For the critical CM, $\hat{k}_c(T)=\frac{3}{8}[(1-\nu)/(1+\nu)](\delta T/\delta T_0)(\epsilon_c/\overline{\epsilon})^2k_c$, where k_c is the "morphological" critical wave vector for misfit $\overline{\epsilon}$ (see Sec. II B).

Above or below $T_c^i(c_0)$, the criteria $\gamma<0$, $2k^{-1}\Gamma^{-1}|\gamma|\epsilon_c^2>1$ also determine the morphological instability of a system which, through some growth process, would have been frozen in a CM state. This could apply to spontaneous kinetic CM's (see Sec. V) or to artificial laterally modulated structures.

B. Surface undulation with half the wavelength of the composition modulation

Equation (16) shows that CM and SU are also coupled in this case. For $T>T_c^i(c_0)$, the system is again stable with respect to both CM and SU. However, the interaction energy is now of order 1 in Δ, as expected from the obvious asymmetry between SU's having opposite signs of Δ. As a consequence, another effect appears: unless $\beta'=0$, the system with a CM of finite amplitude is always unstable with respect to some SU's. $\beta'<0$ induces a SU with crests in the region of extreme deviation from the average composition [$\Delta>0$; Fig. 1(b)], whereas $\beta'>0$ favours troughs in these regions [$\Delta<0$; Fig. 1(c)]. The sign of β' depends on T and on the depth dependence m of the CM. β' can be rewritten as $\beta'=\beta'_{\rm ch}+\beta'_e$, with $\beta'_{\rm ch}=2(1-\nu)Fm^2(t)\delta T/\delta T_0$ and $\beta'_e=2(1+\nu)FP^2(t)$. As mentioned in Sec. III B, $\beta'_{\rm ch}$, favors crests at the CM extrema [Fig. 1(b)] for $T<T_c^i(c_0)$ and troughs [Fig. 1(c)] for $T>T_c^i(c_0)$. On the other hand, since $\beta'_e>0$ (unless the CM is critical, in which case $\beta'_e=0$), the formation of a SU with crests at the CM extrema always increases the elastic energy. For $T>T_c^i(c_0)$, this reinforces the tendency of the alloy not to form zones of extreme composition. However, this increase is simply due to the increased volume of zones of nonaverage composition, which also produces a change in chemical energy. Alternatively, starting from a planar half space, we may compare the changes of elastic energy associated with, respectively, a SU (with crests at the CM extrema) and a perturbation of the CM (without SU) producing the same chemical energy change. We have checked that for a wide range of CM's (including the critical one), the SU costs less elastic energy than the simple CM perturbation. This reflects the easier stress relaxation if crests appear at the CM extrema.

C. The critical compositional modulation and the question of metastability

We have shown that for $\overline{\epsilon}=0$ the static system is thermodynamically stable with respect to CM's above $T>T_c^i$ even if its surface is allowed to undulate with an infinitesimal amplitude. Is this only metastability, or would some surface perturbations of *finite* amplitude reduce the total enthalpy of the system with respect to the homogeneous planar state? No general answer to this question is yet available. In this respect, however, it is interesting to return to the critical CM $m(z)=e^{kz}$, remarkable not only for being the CM with respect to which the planar system becomes thermodynamically unstable against CM at T_c^i, but also because $\sigma_{xx}^p=\sigma_{zz}^p=\sigma_{xz}^p=0$ [from Eq. (4)]. Hence any surface with normal in the *xz* plane is stress free and $\chi=0$. One then easily shows that for any CM amplitude $\eta^{-1}\epsilon_c$ and any free-surface profile $z=t+h(x)$ whose period along x is π/nk with n integer, one has (omitting the positive surface energy term)

$$\delta H(\epsilon_c)=\frac{1}{4\pi}\frac{\delta T}{\delta T_0}\epsilon_c^2\int_0^{2\pi/k}e^{2kh(x)}\sin^2 kx\,dx. \tag{18}$$

The system is thus thermodynamically stable above $T_c^i(c_0)$ with respect to this CM even if its surface is allowed to have finite deviations with respect to planarity.

V. KINETIC INSTABILITY

A. Compositional instability without coupling: Critical growth temperature and optimal modulation

Although apparently pertaining to static layers, the calculations of Sec. III also allow the study of the kinetic instability. We are now interested in the evolution of the alloy upon deposition of extra material (exactly lattice matched on average) on its free surface, assuming that, due to lack of bulk diffusion, the underlying CM remains unaltered. Consider first a half space with CM (3) and planar free surface at $z=t$ $(\Delta=0)$. Since the elastic and chemical energies per unit surface are zero in the reference state, the total enthalpy $H(t)$ of the systems is equal to δH_1 and its rate of change upon addition of a thin alloy layer is $\partial H/\partial t=k^{-1}(\partial\alpha/\partial t)\epsilon_c^2/2$. Since $\partial J_1/\partial t=k[m(t)-J_1(t)]$ and $\partial J_2/\partial t=km^2(t)$, we have

$$\partial H/\partial t = 2(1-\nu)F\left(m^2(t)\,\frac{\delta T}{\delta T_0} + \frac{1+\nu}{1-\nu}[m(t) - 2J_1(t)]^2 \right) k^{-1}\epsilon_c^2.$$

Minimizing this quantity with respect to the depth variation m of the CM amplitude gives the optimum depth variation $\tilde{m}(t)$. Since

$$\partial(\partial H/\partial t)/\partial[m(t)] = 4(1-\nu)F\left(m(t)\,\frac{\delta T}{\delta T_0} + \frac{1+\nu}{1-\nu}[m(t) - 2J_1(t)] \right) k^{-1}\epsilon_c^2,$$

the integral $\tilde{J}_1(t)$ associated with $\tilde{m}$ satisfies $\tilde{J}_1(t) + k^{-1}\partial\tilde{J}_1(t)/\partial t = 2\tilde{J}_1(t)/[1+\phi(T)]$, where $\phi(T) = [(1-\nu)/(1+\nu)]\,\delta T/\delta T_0$. Thus $\tilde{J}_1(t) = \tilde{J}_1(0)\exp(k_z t)$ and $\tilde{m}(t) = \{2\tilde{J}_1(0)/[1+\phi(T)]\}\exp(k_z t)$, with $k_z = \{[1-\phi(T)]/[1+\phi(T)]\}k$. Hence, $\phi(T)=1$ defines a kinetic critical growth temperature $T_c^k(c_0)$ under which pure composition perturbations increase exponentially upon deposition of new material. For $T_c^i(c_0) \leqslant T \leqslant T_c^k(c_0)$, the layer is kinetically but not thermodynamically unstable. We find

$$T_c^k(c_0) = T_c^b(c_0) + 2c_0(1-c_0)\,\frac{\nu}{1-\nu}\,\frac{E\eta^2}{R} = T_c^i(c_0) + c_0(1-c_0)\,\frac{1+\nu}{1-\nu}\,\frac{E\eta^2}{R}, \tag{19}$$

$$k_z = \frac{T_c^k(c_0) - T}{T - T_c^c(c_0)}\,k. \tag{20}$$

The reduced amplification wave number k_z/k is thus a characteristic of the system that depends only on its thermodynamic and elastic properties and on temperature, but not on growth velocity. In particular, $k_z/k = 1$ for $T = T_c^i(c_0)$. Typical variations with temperature and composition for a III-V alloy are represented in Fig. 2.

B. Discussion

We thus recover in a very simple fashion the kinetic critical temperature first calculated by Malyshkin and Shchukin[1] for pure compositional instability (in the elastically anisotropic case) and then by Guyer and Voorhees[15] for coupled instability. Note that our only two hypotheses have already been made by these authors: the continuous selection of the optimal CM amplitude profile $\tilde{m}$ during growth corresponds to fast surface diffusion[1] and the existence of an initial CM of finite amplitude had to be assumed in Refs. 1, 14, and 15. The present results, however, are obtained without explicitly taking deposition fluxes and surface diffusion into account, and in particular without assuming either local vapor/solid equilibrium[15] or step flow growth.[14] Nor did we assume *a priori*[1] that m varies exponentially with z; instead, we calculated the optimum profile. Moreover, the results are obtained for a planar free surface.

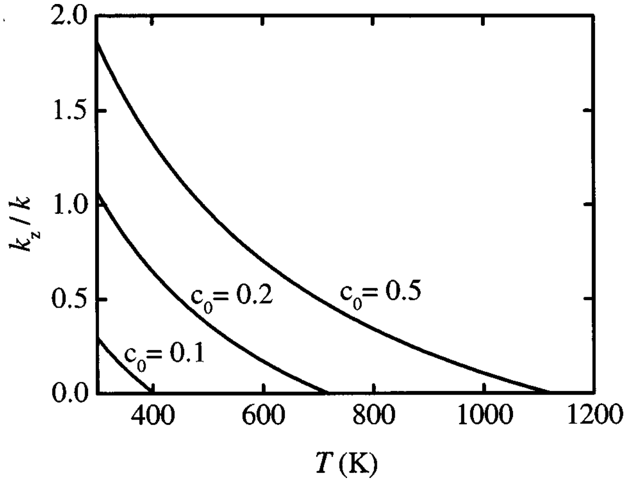

FIG. 2. Variation with temperature of the amplification wave number along growth direction divided by the CM lateral wave number, for various average alloy compositions. Intercepts with horizontal axis give $T_c^k(c_0)$. Material parameters are for $\mathrm{In}_c\mathrm{Ga}_{1-c}$ As alloys (Ref. 3).

This widens considerably the domain of occurrence of the kinetic compositional instability. First, this instability should appear whatever the growth method. CM's have indeed been observed above $T_c^i(c_0)$ in semiconductor alloy layers grown by all the usual epitaxy techniques.[11] Second, Guyer and Voorhees associate the occurrence of the kinetic instability with a coupling between CM and SU; the instability corresponds to a phase shift between CM and SU and the CM amplitude is explicity coupled to the SU amplitude (proportional to it when $\Gamma = 0$). We demonstrate here that the kinetic composition instability does not require any morphological surface perturbation to develop. Because of this and because it applies as long as linear elasticity holds, the present calculation is also not limited to the initial stage of instability development. On the other hand, the calculations of Sec. III, which take explicitly into account nonplanar free surfaces, also extend the domain of occurrence of the compositional kinetic instability with respect to the work of Malyshkin and Shchukin,[1] who assumed a planar surface. Indeed, the results of Sec. V A are still valid for a nonplanar layer with a SU amplitude remaining constant during growth (the reference-state energy is then constant). Hence, layers with corrugated surfaces are also kinetically unstable with respect to CM's for $T < T_c^k(c_0)$.

Finally, we may consider the evolution of small-amplitude SU's during growth. For a sinusoidal SU having the same wave vector as the CM, an increase of amplitude produces a change of the average composition of either the newly deposited or the already deposited material, which thereby deviates from strict lattice matching (as previously noticed[2] and as appears from the proportionality of W_c^{ref} to Δ). Although probably important in practice,[12,13] the former case is out of the scope of the present study, whereas the latter corresponds to thermodynamical instability (Sec. IV). SU amplification is, however, compatible with lattice matching for SU's having half the wavelength of the CM. Then, $\partial H/\partial\Delta = \beta'(t)\epsilon_c^2$ and the discussion of the morphological instability in Sec. IV B shows that the surface perturbation will tend to develop once a finite-amplitude CM has appeared, which is anyway also a prerequisite for the development of the kinetic compositional instability. For the optimal CM $\tilde{m}$, $\beta'(t)>0$ in the domain of pure kinetic compositional instability ($T_c^i \leqslant T \leqslant T_c^k$), so that we expect half-wavelength SU's with troughs in the regions of extreme deviation from average composition. That only coupling between perturbations with identical wavelengths seems to have been actually observed so far might be related to the residual lattice mismatch of the layers. To summarize, both instabilities should develop together, although a finite CM is needed for the SU to appear whereas no SU is needed for the CM to grow.

VI. SUMMARY AND CONCLUSIONS

We performed an analytical calculation of the elastic relaxation of a lateral composition modulation coupled to a surface undulation of equal or half wavelength in a lattice-matched epitaxial alloy layer or alloy half space. The coupling energy between the two perturbations, calculated up to second order in undulation amplitude, is nonzero in both cases. These calculations allowed us to treat in a unified fashion the instabilities affecting the compositional homogeneity as well as the surface planarity of the layer, whether thermodynamic (in the case of a closed system) or kinetic (for an open system). In particular, we determined how these perturbations condition or affect each other. In sharp contrast to the case of lattice-mismatched layers, the thermodynamic critical temperature T_c^i below which the static layer becomes unstable with respect to composition variations is identical for corrugated layers and for planar ones. The kinetic critical temperature T_c^k, below which composition modulations get amplified during growth, lies well above T_c^i. It is an intrinsic property of the system independent of any particular growth method or growth mode. The development of the kinetic composition modulations does not require any surface nonplanarity. However, because of the aforementioned coupling, a layer where such a composition modulation has started to develop becomes in turn unstable with respect to surface undulations. Whereas the details of the development or inhibition of the kinetic instability can probably be obtained only through consideration of the particular processes operating in given growth conditions,[14–16] our calculations underline the universal character of the instability, independent of these details.

APPENDIX A

Stresses are calculated to second order in Δ on the free surface $z=t+h(x)$, on which the boundary conditions are $(\sigma_{xx}^p+\sigma_{xx}^A)h'(x)-(\sigma_{xz}^p+\sigma_{xz}^A)=0$, $(\sigma_{xz}^p+\sigma_{xz}^A)h'(x)-(\sigma_{zz}^p+\sigma_{zz}^A)=0$. With terms up to $n=3$ included in the Airy function χ, whose coefficients are at least of order 1 in Δ,

$$\sigma_{xx}^A h'(x)-\sigma_{xz}^A=k\sum_{n=1}^{4} c_n^1\cos nkx+s_n^1\sin nkx,$$

$$\sigma_{xz}^A h'(x)-\sigma_{zz}^A=k\sum_{n=1}^{4} c_n^2\cos nkx+s_n^2\sin nkx \qquad \text{(A1)}$$

with

$$c_1^1=B_1+kA_1-2k\Delta(B_2'+kA_2'),$$

$$c_1^2=kA_1'+k\Delta(B_2+2kA_2),$$

$$c_2^1=2B_2+4kA_2+k\Delta(2B_1'+kA_1'-6B_3'-9kA_3'),$$

$$c_2^2=4kA_2'-k\Delta(B_1+kA_1-3B_3-9kA_3),$$

$$c_3^1=3[B_3+3kA_3+2k\Delta(B_2'+kA_2')],$$

$$c_3^2=3[3kA_3'-k\Delta(B_2+2kA_2)],$$

$$c_4^1=6k\Delta(2B_3'+3kA_3'),\quad c_4^2=-6k\Delta(B_3+3kA_3).$$

s_n^j is obtained from c_n^j by exchanging A_i and A_i' and B_i and B_i', respectively, and multiplying each coefficient by $(-1)^{j+n+i}$. Then

$$I(k,t+h(x))=I(k,t)+P(t)e^{-kt}h(x)$$
$$+k[Q(t)-P(t)]e^{-kt}h^2(x)+O(\Delta^3),$$

$$G(k,t+h(x),t+\Delta)=[\Delta-h(x)]m(t)e^{-kt}+kQ(t)e^{-kt}$$
$$\times[\Delta^2-h^2(x)]+O(\Delta^3),$$

$$m(t+h(x))=m(t)+h(x)m'(t)+m''(t)h^2(x)/2+O(\Delta^3), \qquad \text{(A2)}$$

where $P(t)=m(t)-2kI(k,t)e^{kt}$ and $Q(t)=[m'(t)-km(t)]/2k$. Consequently,

$$\sigma_{xx}^p h'(x)-\sigma_{xz}^p=F\epsilon_c k\Delta(\{8P(t)-k\Delta[15P(t)-2Q(t)]\}$$
$$\times\cos kx+3k\Delta[P(t)+2Q(t)]$$
$$\times\cos 3kx-2P(t)(1-2k\Delta)\sin 2kx),$$

$$\sigma_{xz}^p h'(x)-\sigma_{zz}^p=-F\epsilon_c k^2\Delta^2(8\cos 2kx+5\sin kx$$
$$-3\sin 3kx)P(t).$$

Writing the boundary conditions in terms of the Airy function coefficients, we readily find that $A_2, B_2, A_1', B_1', A_3', B_3'$ are of order 3 in Δ at least. The six remaining coefficients satisfy the following system, whose solution is given by Eq. (7):

$$B_1+kA_1-2k\Delta B_2=-F\epsilon_c\{8P(t)$$
$$+k\Delta[2Q(t)-15P(t)]\}\Delta,$$

$$B_3+3kA_3+2k\Delta B_2'=-F\epsilon_c[P(t)+2Q(t)]k\Delta^2,$$

$$-2B_2'-4kA_2'+2k\Delta B_1=8F\epsilon_c P(t)(1-2k\Delta)\Delta,$$

$$(4A_2'-\Delta B_1)/8=(A_1-\Delta B_2')/5=-(3A_3+\Delta B_2')$$
$$=F\epsilon_c P(t)\Delta^2. \qquad \text{(A3)}$$

APPENDIX B

With terms up to $n=5$ in χ, Eq. (A1) still holds with now terms up to $n=7$ in the sum and

$$c_1^1 = B_1 + kA_1 + k\Delta(2B_1' + kA_1' - 6B_3' - 9kA_3')/2,$$

$$c_1^2 = kA_1' - k\Delta(B_1 + kA_1 - 3B_3 - 9kA_3)/2,$$

$$c_2^1 = 2[B_2 + 2kA_2 - 4k\Delta(B_4' + 2kA_4')],$$

$$c_2^2 = 4[kA_2' + k\Delta(B_4 + 4kA_4)],$$

$$c_3^1 = 3[B_3 + 3kA_3 + k\Delta(2B_1' + kA_1' - 10B_5' - 25kA_5')/2],$$

$$c_3^2 = 3[3kA_3' - k\Delta(B_1 + kA_1 - 5B_5 - 25kA_5)/2],$$

$$c_4^1 = 4[B_4 + 4kA_4 + 2k\Delta(B_2' + kA_2')],$$

$$c_4^2 = 4[4kA_4' - k\Delta(B_2 + 2kA_2)],$$

$$c_5^1 = 5[B_5 + 5kA_5 + 3k\Delta(2B_3' + 3kA_3')/2],$$

$$c_5^2 = 5[5kA_5' - 3k\Delta(B_3 + 3kA_3)/2],$$

$$c_6^1 = 24k\Delta(B_4' + 2kA_4'), \quad c_6^2 = -12k\Delta(B_4 + 4kA_4),$$

$$c_7^1 = 35k\Delta(2B_5' + 5kA_5')/2, \quad c_7^2 = -35k\Delta(B_5 + 5kA_5)/2.$$

s_n^j is obtained from c_n^j by exchanging A_i and A_i' and B_i and B_i', respectively, and multiplying each coefficient by $(-1)^{j+n'+i'}$ where $n' = n/2$ for n even, $n' = (n-1)/2$ for n odd, $i' = i/2$ for i even, and $i' = (i-1)/2$ for i odd. Moreover, on the free surface $z = t + h(x)$,

$$\begin{aligned}\sigma_{xx}^p h' - \sigma_{xz}^p = 2F\epsilon_c k\Delta\big(&\{2P(t) - k\Delta[P(t) - 2Q(t)]\}(\cos kx\\ &- \sin kx) - \{6P(t) - k\Delta[10P(t) - 3Q(t)]\}\\ &\times(\cos 3kx + \sin 3kx) + k\Delta[2P(t) + 5Q(t)]\\ &\times(\cos 5kx - \sin 5kx)\big),\end{aligned}$$

$$\begin{aligned}\sigma_{xz}^p h' - \sigma_{zz}^p = 5F\epsilon_c k^2\Delta^2 P(t)[&-2(\cos kx + \sin kx)\\ &-3(\cos 3kx + \sin 3kx) + \cos 5kx + \sin 5kx].\end{aligned}$$

We find that $A_2, B_2, A_4, B_4, A_2', B_2', A_4', B_4'$ are of order 3 at least and the 12 remaining coefficients satisfy the following system, whose solution is given by Eq. (14):

$$\begin{aligned}&B_1 + kA_1 + k\Delta B_1' - 3k\Delta B_3'\\ &\quad = B_1' + kA_1' + k\Delta B_1 + 3k\Delta B_3\\ &\quad = -4F\epsilon_c\{P(t) - k\Delta[P(t) - Q(t)]\},\\[1ex] &3B_3 + 9kA_3 + 3k\Delta B_1'\\ &\quad = -(3B_3' + 9kA_3' - 3k\Delta B_1)\\ &\quad = 2F\epsilon_c k\Delta\{6P(t) - k\Delta[10P(t) - 3Q(t)]\},\\[1ex] &-(15k\Delta B_3' + 5B_5 + 25kA_5)\\ &\quad = 15k\Delta B_3 - 5B_5' - 25kA_5'\\ &\quad = 2F\epsilon_c k\Delta^2[2P(t) + 5Q(t)],\\[1ex] &-2A_1' + \Delta B_1 - 3\Delta B_3\\ &\quad = -2A_1 + \Delta B_1' + 3\Delta B_3' = -20F\epsilon_c P(t)\Delta^2,\\[1ex] &6A_3' - \Delta B_1 = -(6A_3 + \Delta B_1') = 10F\epsilon_c P(t)\Delta^2,\\[1ex] &3\Delta B_3 - 10A_5' = -(3\Delta B_3' + 10A_5) = 2F\epsilon_c P(t)\Delta^2.\end{aligned} \tag{B1}$$

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