Curvature contributions to the capillary-wave Hamiltonian for a pinned interface

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The curvature contributions to the capillary-wave Hamiltonian of a pinned interface are analyzed within the mean-field version of the Landau-Ginzburg-Wilson theory supplemented by the crossing constraint. The resulting fourth-order Hamiltonian can be unambiguously written in the Helfrich form with the coefficients depending on the local distance l of the fluctuating interface from the flat substrate. The expressions for these coefficients are derived and their l dependence is discussed; they all consist of exponentially decaying terms multiplied by polynomials. The expression for the l-dependent stiffness coefficient present in the fourth-order Hamiltonian differs from one derived recently within a second-order theory.

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I. INTRODUCTION

The structure of the capillary-wave Hamiltonian, which represents the cost in free energy to deform the flat interface into a given rippled configuration, has been intensively studied in recent years [1–43]. In these studies, mostly devoted to free interfaces, special emphasis has been put on the form of the functional dependence of the capillary-wave Hamiltonian on the interfacial configuration. This dependence plays an important role, e.g., in the interpretation of the scattering experiments [44–51]. If, however, one studies wetting [3,4,8,9,11,12,14,19,20,32,35,36,38,40–43], then the knowledge of the capillary-wave Hamiltonian for a *pinned* interface is essential. Recently the wetting transitions in three-dimensional systems with short-range forces, for which the upper critical dimension is 3, have received special attention because of the nonuniversal indices characterizing these transitions [3,8,9,19,20,30–32,35–38]. As the paradigm of a system undergoing the wetting transition one usually considers the uniaxial ferromagnet in which the magnetization $m(z,\boldsymbol{R})$ serves as the order parameter; z measures the distance from the substrate surface and $\boldsymbol{R}=(x,y)$. Until now the studies of this problem [3,8,11,12,20,30,32,35] were concentrated on the leading term in the capillary-wave Hamiltonian. Indeed, Jin and Fisher [32] showed recently that for a fluctuating interface separating the bulk phase α from the β-like phase adsorbed on a flat substrate the leading contribution to the capillary-wave Hamiltonian has the form

$$\mathcal{H}_{\rm CW}[l]=\int d\boldsymbol{R}\left[\frac{1}{2}\,\Sigma_{\alpha\beta}(l)\,(\nabla l)^2\,+\,V(l)\right]\,, \qquad (1)$$

where $l=l(\boldsymbol{R})$ denotes the actual interfacial configuration, i.e., the local distance of the fluctuating α-β interface from the substrate surface. $V(l)$ is the effective interface potential describing the interaction of the α-β interface with the substrate. The important feature of Eq. (1) is that it contains the l-dependent stiffness coefficient $\Sigma_{\alpha\beta}(l)$, which replaces the constant stiffness $\sigma_{\alpha\beta}$ of the *free* α-β interface considered in previous studies [3,4,8,11,14]. It has been shown that $\Sigma_{\alpha\beta}(l)$ contains, in addition to its asymptotic value $\Sigma_{\alpha\beta}(\infty)=\sigma_{\alpha\beta}$, the exponentials multiplied by the polynomials, i.e., terms $w_{jk}\,l^k\,e^{-j\kappa l}$, where $0\le k\le j=1,2,\ldots$ and $\kappa=\xi_\beta^{-1}$ is the inverse correlation length in the bulk phase β. The presence of the polynomials multiplying the exponentials turned out to be crucial in the renormalization-group analysis of the order of the wetting transition; for an extensive discussion and comparison with other approaches see [32].

Equation (1) contains only the leading-order term $[\sim(\nabla l)^2]$ in the gradient expansion of the complete capillary-wave Hamiltonian. This leading-order term can be classified as being of second order in the spatial derivatives; indeed, the sum of the orders of the derivatives of l with respect to x or y is equal to 2 in this term. However, another term of second order could, at least in principle, be present in Eq. (1). This is a term proportional to Δl, where Δ denotes the two-dimensional Laplacian. This would be the mean curvature term because up to the second-order terms the mean curvature $H=\frac{1}{2}\Delta l$; see below. It was shown in our previous work [39] that, although in the process of systematic derivation of all the second-order terms contributing to the capillary-wave Hamiltonian the terms proportional to Δl do appear, their sum is identically zero and the only remaining contribution is the one present in Eq. (1).

For *free* interfaces the capillary-wave Hamiltonian has, up to the curvature terms, the Helfrich form [1]

$$\mathcal{H}^0_{\rm CW}[l]=\int d\boldsymbol{R}\Big\{\sigma_{\alpha\beta}\,\Big[\sqrt{1+(\nabla l)^2}\,-\,1\Big] + \sqrt{1+(\nabla l)^2}\,\left[c^0_H\,H\,+\,c^0_{H^2}\,H^2\,+\,c^0_G\,G\right]\Big\}, \qquad (2)$$

where

$$H = \frac{l_{xx}(1+l_y^2) + l_{yy}(1+l_x^2) - 2l_{xy}l_x l_y}{2(1+l_x^2+l_y^2)^{\frac{3}{2}}} \tag{3}$$

is the mean curvature and

$$G = \frac{l_{xx}l_{yy} - l_{xy}^2}{(1+l_x^2+l_y^2)^2} \tag{4}$$

is the Gaussian curvature. Up to the second-order terms the above capillary-wave Hamiltonian [Eq. (2)] reduces the form

$$\mathcal{H}^0_{\rm CW}[l] = \int d\boldsymbol{R} \left\{ \frac{1}{2}\,\sigma_{\alpha\beta}\,(\nabla l)^2 + \frac{1}{2}\,c_H^0\,\Delta\, l \right\}. \tag{5}$$

For smooth and asymptotically flat interfaces the second term in Eq. (5) vanishes and one is left with the standard form of the second-order capillary-wave Hamiltonian containing $(\nabla l)^2$ only. This is not the case for a pinned interface. Suppose that in this case the second-order capillary-wave Hamiltonian has again the structure given by Eq. (5) but with the l-dependent coefficients $\Sigma_{\alpha\beta}(l)$ and $\mathcal{C}_H(l)$; it also contains the effective interface potential $V(l)$. An arbitrary part of the mean curvature term $\frac{1}{2}\int d\boldsymbol{R}\,\mathcal{C}_H(l)\Delta l$, namely, $\frac{1}{2}\int d\boldsymbol{R}\,\mathcal{C}_H(l)\,\mathcal{A}(l)\,\Delta l$, where $\mathcal{A}(l)$ is an arbitrary smooth function that derivative vanishes at infinity, can be integrated by parts into $-\frac{1}{2}\int d\boldsymbol{R}\,[\mathcal{A}(l)\mathcal{C}_H(l)]'\,(\nabla l)^2$, where the prime denotes the derivative with respect to l; the remaining part, i.e., $\frac{1}{2}\int d\boldsymbol{R}\,[1-\mathcal{A}(l)]\,\mathcal{C}_H(l)\,\Delta l$, still represents the mean curvature contribution. The above procedure of splitting the integrand and integration by parts results in the modification of the coefficient multiplying $(\nabla l)^2$. Even imposing the boundary condition that the limiting value (at $l=\infty$) of the coefficients multiplying $\frac{1}{2}(\nabla l)^2$ and Δl have to be equal to the known coefficient $\sigma_{\alpha\beta}$ and c_H^0 for free interfaces does not remove this ambiguity because $\mathcal{C}'_H(\infty) = \mathcal{A}'(\infty) = 0$. Thus there exists certain ambiguity in the structure of the l-dependent coefficients present in the second-order capillary-wave Hamiltonian for a pinned interface.

In this paper we show that *up to fourth-order terms* the capillary-wave Hamiltonian for a pinned interface can be *unambigously* written as

$$\begin{aligned}\mathcal{H}_{\rm CW}[l] = \int d\boldsymbol{R} \Big\{ & \Sigma^*_{\alpha\beta}(l)\,\left[\sqrt{1+(\nabla l)^2}\,-1\right] \\ & + \sqrt{1+(\nabla l)^2}\,[\mathcal{C}^*_H(l)\,H \\ & + \mathcal{C}^*_{H^2}(l)\,H^2 + \mathcal{C}^*_G(l)\,G] + V(l) \Big\}\,,\end{aligned} \tag{6}$$

and we derive the explicit expressions for the l-dependent coefficients $\Sigma^*_{\alpha\beta}(l), \mathcal{C}^*_H(l), \mathcal{C}^*_{H^2}(l), \mathcal{C}^*_G(l)$. In the limit $l \to \infty$ these coefficients reduce to the values we derive by an independent method for the free case.

The interesting conclusion resulting from Eq. (6) is that the coefficient of the surface stiffness $\Sigma^*_{\alpha\beta}(l)$ as derived by us within the fourth-order theory is different from the one derived recently within the second-order theory. In principle, this difference may lead to new predictions (based on the functional renormalization) for the wetting transitions. However, we show in Sec. IV that the renormalization of the capillary-wave Hamiltonian given by Eq. (6) but including terms up to second-order only leads to the same conclusions as the standard theory. New predictions may come out when the capillary-wave Hamiltonian is renormalized up to fourth-order terms. This will be the subject of our further work.

In Sec. II we present a systematic way to derive the expression for the constrained magnetization profile up to an arbitrary order. This profile is then used to derive the expression for the constrained magnetization profile up to an arbitrary order. This profile is then used to derive the expression for the capillary-wave Hamiltonian, which includes terms of arbitrary order.

In Sec. III we concentrate on the fourth-order terms and we derive the analog of the Helfrich Hamiltonian for the pinned interface and obtain explicit expressions for the l-dependent surface stiffness and for the l-dependent coefficients multiplying the curvature terms. Section IV contains a discussion of the results.

II. GENERAL EXPRESSION FOR THE CONSTRAINED MAGNETIZATION PROFILE AND FOR THE CAPILLARY-WAVE HAMILTONIAN

To derive the constrained magnetization profile and the capillary-wave Hamiltonian we employ the mean-field approach, starting from the Landau-Ginzburg-Wilson (LGW) Hamiltonian for a semi-infinite ($z \geq 0$) uniaxial ferromagnet in the presence of a flat substrate

$$\begin{aligned}\mathcal{H}_{\rm LGW}[m] = \int d\boldsymbol{R} \Bigg(\int_0^\infty dz \Bigg\{ & \frac{K}{2}\left[\left(\frac{\partial m}{\partial z}\right)^2 + (\nabla m)^2\right] \\ & + \Phi_0(m) \Bigg\} + \Phi_1(m|_{z=0}) \Bigg).\end{aligned} \tag{7}$$

The bulk free-energy density $\Phi_0(m)$ is taken in the form of the double parabola [6,32,39] having two equal minima at $m_{\alpha 0} < 0$ and $m_{\beta 0} > 0$ (thermodynamically the system is located at the α-β coexistence line)

$$\Phi_0(m) = \begin{cases} \frac{Kp_\alpha^2}{2}\,(m\,-\,m_{\alpha 0})^2 & \text{for } m \leq 0 \\ \frac{Kp_\beta^2}{2}\,(m\,-\,m_{\beta 0})^2 & \text{for } m \geq 0, \end{cases} \tag{8}$$

where p_α, p_β are the inverse bulk correlation lengths for the α and β phases, respectively. This simple form of $\Phi_0(m)$ allows for the analytical derivation of the expressions for the magnetization and for the capillary-wave Hamiltonian. The surface free-energy density

$$\Phi_1(m_1) = -h_1\,m_1\,-\,\frac{1}{2}gm_1^2\,, \tag{9}$$

where $m_1 = m(z=0, \boldsymbol{R})$, contains the surface field h_1 whose positive values stabilize the β-like phase near the substrate. The LGW Hamiltonian is supplemented by the constraint defining the position of the interface. Here we follow Jin and Fisher and choose the crossing criterion [31,32,36,37,39] according to which the value of the magnetization at the interface, i.e., at $z = l$, is equal to zero. The profile l is assumed to be asymptotically flat, i.e., for $R \to \infty$, l tends to a constant (l_π) and all its spatial derivatives tend to zero. Within the mean-field approach the magnetization profile including the α-β interface is obtained via minimization of the LGW Hamiltonian under the "crossing" constraint. In this way one obtains the constrained magnetization $m(z, \boldsymbol{R}; [l])$, which is a functional of the interfacial shape l and fulfills the crossing constraint

$$m(z = l, \boldsymbol{R}; [l]) = 0 \; . \tag{10}$$

The continuity of $\Phi_0(m)$ at the crossing value ($m^\times = 0$) gives

$$\frac{p_\alpha}{p_\beta}\frac{m_{\alpha 0}}{m_{\beta 0}} = -1 \; . \tag{11}$$

The constrained order parameter is denoted in phases α and β, i.e., for $z \geq l$ and for $0 \leq z \leq l$, by $m_\alpha = m_\alpha(z, \boldsymbol{R}; [l])$ and $m_\beta = m_\beta(z, \boldsymbol{R}; [l])$, respectively. The minimization of the LGW Hamiltonian leads to the following equation for the constrained order parameter:

$$\left(\frac{\partial^2}{\partial z^2} + \Delta\right) m_\gamma = p_\gamma^2 \, (m_\gamma - m_{\gamma 0}) \; , \tag{12}$$

where $\gamma = \alpha, \beta$. Equation (12) is supplemented by the boundary conditions at the substrate's surface and at infinity:

$$K \left.\frac{\partial m_\beta(z, \boldsymbol{R}; [l])}{\partial z}\right|_{z=0} = -h_1 - g\, m_\beta(z = 0, \boldsymbol{R}; [l]) \; , \tag{13}$$

$$\lim_{z\to\infty} m_\alpha = m_{\alpha 0} \; . \tag{14}$$

Our method of deriving the fourth-order capillary-wave Hamiltonian can also be applied to the case of the *extended* LGW theory, which contains, on the right-hand side (rhs) of Eq. (7), the *extra* term $B\left(\frac{\partial^2 m}{\partial z^2} + \Delta m\right)^2$, $B = \text{const}$ [24,27]. The extended theory also leads to a linear partial-differential equation for the constrained magnetization, which can be solved with the help of our procedure. One expects then the B-proportional contributions to the surface stiffness as well as to the coefficients multiplying the curvatures; see [24] for an extended discussion of this problem for a spherical interface.

A. Constrained magnetization

The solution of Eqs. (10) and (12)–(14) is obtained separately in each region defined by the double-parabola potential. We restrict our presentation to the β phase, where the analysis is more involved; in the α phase the analysis proceeds similarly to the free case.

First we recall the solution up to the second-order terms, which was obtained previously [39]:

$$m_\gamma(z, \boldsymbol{R}; [l]) = m_{\pi\gamma}(z\,; l) + b_{1\gamma}(z\,; l)\, \Delta\mathcal{M}_\gamma \; , \tag{15}$$

where

$$\begin{aligned} m_{\pi\alpha}(z; l) &= m_{\alpha 0}\,\{1 - \exp[p_\alpha(l - z)]\} \; , \\ m_{\pi\beta}(z\,; l) &= m_{\beta 0} + B_+ \exp[p_\beta(z - l)] \\ &\quad + B_- \exp[p_\beta(l - z)] \; , \end{aligned} \tag{16}$$

and

$$B_+ = -\frac{m_{\beta 0} + \tau X}{1 - \mathcal{G}X^2} \; , \; B_- = \frac{\tau + m_{\beta 0} X \mathcal{G}}{1 - \mathcal{G}X^2} X,$$
$$X = \exp(-p_\beta l) \; , \; \tau = \frac{h_1 + g m_{\beta 0}}{K p_\beta - g} \; , \; \mathcal{G} = -\frac{K p_\beta + g}{K p_\beta - g} \; . \tag{17}$$

The functions $b_{1\gamma}$ and $\mathcal{M}_\gamma$ are given by

$$b_{1\alpha}(z\,; l) = \frac{z - l}{2 p_\alpha} \exp(-p_\alpha z) \; , \tag{18}$$

$$\begin{aligned} b_{1\beta}(z\,; l) = \frac{1}{p_\beta}\Bigg\{ &\frac{l - z}{2} \exp(p_\beta z) - \mathcal{G}\, \frac{l + z}{2} \exp(-p_\beta z) \\ &+ \frac{\mathcal{G} l - p_\beta \rho}{1 - \mathcal{G}X^2} X \exp[p_\beta(z - l)] \\ &- \frac{\mathcal{G}^2 X^2 l - p_\beta \rho}{1 - \mathcal{G}X^2} X \exp[-p_\beta(z - l)] \Bigg\} \; , \end{aligned}$$

where $\rho = g\,K/[p_\beta(K p_\beta - g)^2]$ and

$$\mathcal{M}_\alpha = -m_{\alpha 0} \exp(p_\alpha l) \; , \; \mathcal{M}_\beta = -\frac{m_{\beta 0} + \tau X}{1 - \mathcal{G}X^2} X \; . \tag{19}$$

τ [Eq. (17)] measures the distance from the critical wetting point along the coexistence line $\tau \sim T - T_w$.

In view of the above results we look for the general solution of Eqs. (10) and (12)–(14) in the form

$$m_\beta(z, \boldsymbol{R}; [l]) = m_{\beta 0} + \tau \exp(-p_\beta z) + F_\beta(z, \Delta)\, M_\beta \; , \tag{20}$$

where $F_\beta(z, \Delta)$ is a z-dependent pseudodifferential operator and M_β is a function of $\boldsymbol{R}$ depending on l, Δl, and $(\nabla l)^2$. Equation (20) is derived by writing the solution of Eq. (12) as

$$m_\beta(z, \boldsymbol{R}; [l]) = m_{\beta 0} + \exp(z\sqrt{p_\beta^2 - \Delta})\, M_\beta^1 + \exp(-z\sqrt{p_\beta^2 - \Delta})\, M_\beta^2 \; , \tag{21}$$

where

$$M^i_\beta(\boldsymbol{R};\,[l]) = \int \frac{d\boldsymbol{k}}{(2\pi)^2} \exp[(i\boldsymbol{k}\cdot\boldsymbol{R})]\ \mathcal{F}^i_\beta(\boldsymbol{k};\,[l])$$

$$i = 1,2\,. \quad (22)$$

$\mathcal{F}^i_\beta(\boldsymbol{k};\,[l])$ are arbitrary functions such that the crossing criterion is fulfilled. Then, with the help of the crossing criterion, one expresses M^2_β as a functional of M^1_β, which finally leads to Eq. (20); see below. Indeed, it was shown previously [39] that *up to the second-order terms*

$$\begin{aligned} m_\beta(z,\boldsymbol{R};[l]) \simeq{}& m_{\beta 0} + \tau\exp(-p_\beta z) \\ &+\left\{\exp(p_\beta z)\left(1-\frac{z\Delta}{2p_\beta}\right)\right. \\ &\left. -\exp(p_\beta z)\left[\mathcal{G}+\left(\frac{z\mathcal{G}}{2p_\beta}-\rho\right)\Delta\right]\right\}M^1_\beta\,; \end{aligned} \quad (23)$$

after deriving the expression for M^1_β and inserting it into Eq. (23) this equation reduces to Eqs. (15)–(19).

In the present analysis we aim at finding the magnetization profile and the capillary-wave Hamiltonian up to an arbitrary order. For this purpose the yet unknown functions F_β and M_β in Eq. (20) are represented as the formal power series

$$F_\beta(z,\,\Delta) = \sum_{n=0}^{\infty} F_{n\beta}(z)\,\Delta^n \quad (24)$$

and

$$M_\beta = \sum_{n=0}^{\infty} M_{n\beta}\,. \quad (25)$$

$M_{n\beta}$ represents that part of M_β which contains all spatial derivatives of l of orders that add up to $2n$; it is thus a linear combination of terms $(\Delta l)^i\,[(\nabla l)^2]^{n-i}$ with l-dependent coefficients. The above expansions (24) and (25) lead to the expression for m_β

$$m_\beta(z,\,\boldsymbol{R};\,[l]) = \sum_{n=0}^{\infty} m_{n\beta}(z,\,\boldsymbol{R};\,[l]), \quad (26)$$

where

$$m_{0\beta} = m_{\pi\beta}(z\,;l)\,,$$

$$m_{1\beta} = F_{0\beta}(z)\,M_{1\beta} + F_{1\beta}(z)\,\Delta M_{0\beta}, \quad (27)$$

$$\begin{aligned} m_{2\beta} ={}& F_{0\beta}(z)\,M_{2\beta} + F_{1\beta}(z)\,\Delta M_{1\beta} \\ &+F_{2\beta}(z)\,\Delta^2 M_{0\beta}\,,\ \ldots\,. \end{aligned}$$

Generally,

$$\begin{aligned} m_{0\beta} &= m_{\beta 0} + \tau\exp(-p_\beta z) + F_{0\beta}(z)\,M_{0\beta}, \\ m_{n\beta} &= \sum_{i=0}^{n} F_{i\beta}(z)\,\Delta^i\,M_{n-i\,\beta}, \quad n \ge 1 \end{aligned} \quad (28)$$

and we demand the crossing criterion to be fulfilled in each order

$$m_{n\beta}(z = l,\,\boldsymbol{R};\,[l]) = 0\,. \quad (29)$$

In our previous work [39] we determined $m_{0\beta}$ and $m_{1\beta}$. The knowledge of these terms was sufficient to calculate the surface stiffness coefficient. This algorithm can be continued to an arbitrary order, though for $n > 2$ the analysis becomes cumbersome. One obtains the expressions for $M_{n\beta}$, $n = 0, 1, \ldots$,

$$M_{0\beta} = -\frac{m_{\beta 0} + \tau X}{F_{0\beta}(l)}\,,$$

$$M_{1\beta} = -\frac{F_{1\beta}(l)}{F_{0\beta}(l)}\,\Delta M_{0\beta}, \quad (30)$$

$$M_{2\beta} = -\frac{F_{1\beta}(l)}{F_{0\beta}(l)}\,\Delta M_{1\beta} - \frac{F_{2\beta}(l)}{F_{0\beta}(l)}\,\Delta^2 M_{0\beta}, \ldots\,.$$

Generally, for $n > 0$, one has

$$M_{n\beta} = -\sum_{i=1}^{n} \frac{F_{i\beta}(l)}{F_{0\beta}(l)}\,\Delta^i M_{n-i\beta}\,. \quad (31)$$

After substituting $M_{n\beta}$ [Eq. (31)] into Eq. (28), one obtains

$$m_{n\beta} = \sum_{i=1}^{n} b_{i\beta}(z;\,l)\,\Delta^i M_{n-i\beta}\,, \quad (32)$$

where

$$b_{i\beta}(z\,;l) = F_{i\beta}(z) - F_{i\beta}(l)\,\frac{F_{0\beta}(z)}{F_{0\beta}(l)}\,. \quad (33)$$

Note that for all i, $b_{i\beta}(z = l\,;l) = 0$ and thus the crossing criterion is fulfilled in every order. The functions $F_{i\beta}(z)$ remain to be determined in the process of constructing, order by order, the solution m_β [Eqs. (26), (30), and (33)].

B. Capillary-wave Hamiltonian

In this section we sketch the derivation of the general expression for the capillary-wave Hamiltonian that includes terms of arbitrary order; details are presented in Appendix A. For this purpose each of the four terms $\mathcal{H}_{i\beta}$, $i = 1, \ldots, 4$, representing the different β-phase contributions to the LGW Hamiltonian [Eq. (7)], i.e.,

$$\mathcal{H}_{\beta 1} = \frac{K}{2}\int d\boldsymbol{R}\int_0^l dz\,\left(\frac{\partial m_\beta}{\partial z}\right)^2\,, \quad (34)$$

$$\mathcal{H}_{\beta 2} = \frac{K}{2}\int d\boldsymbol{R}\int_0^l dz\,\left(\nabla m_\beta\right)^2\,, \quad (35)$$

$$\mathcal{H}_{\beta 3} = \frac{K p_\beta^2}{2} \int d\boldsymbol{R} \int_0^l dz \ (m_\beta - m_{\beta 0})^2 \ , \qquad (36)$$

$$\mathcal{H}_{\beta 4} = - \int d\boldsymbol{R} \left(h_1 m_1 + \frac{1}{2} g m_1^2 \right) \ , \qquad (37)$$

is split, with the help of Eq. (26), into

$$\mathcal{H}_{i\beta} = \sum_{n=0}^{\infty} \mathcal{H}_{i\beta}^{(n)}, \quad i = 1, \ldots, 4 \ , \qquad (38)$$

where $\mathcal{H}_{i\beta}^{(n)}$ represents the $2n$th-order contribution to $\mathcal{H}_{i\beta}$. Each contribution $\mathcal{H}_{i\beta}^{(n)}$, $i = 1, \ldots, 4$, is then analyzed separately. After integrating by parts and using the crossing criterion [Eq. (29)] and Eqs. (12) and (13) the resulting expression for $\mathcal{H}_\beta$ can be written in a transparent form

$$\mathcal{H}_\beta = \int d\boldsymbol{R} \left[-\frac{K}{2} \int_0^l dz \, m_{\pi\beta} \, \Delta \, m_\beta + V(l) \right] . \qquad (39)$$

$V(l)$ in Eq. (39) represents the effective interface potential [29–31]

$$V(l) = \sigma_{\alpha\beta} + \sigma_{w\beta} + \mathcal{V}(l) \ , \qquad (40)$$

where $\sigma_{w\beta}$ is the surface stiffness coefficient of the wall-β interface and $\sigma_{\alpha\beta}$ is the surface stiffness coefficient for a free α-β interface,

$$\sigma_{w\beta} = \frac{K}{2} p_\beta \tau^2 - h_1(\tau + m_{\beta 0}) - \frac{1}{2} g (\tau + m_{\beta 0})^2 \ , \qquad (41)$$

$$\sigma_{\alpha\beta} = \frac{K}{2} \left(m_{\alpha 0}^2 \, p_\alpha + m_{\beta 0}^2 \, p_\beta \right) \ , \qquad (42)$$

and

$$\mathcal{V}(l) = \frac{v_1 X + v_2 X^2}{1 - \mathcal{G} X^2} \ ; \qquad (43)$$

$v_1 = 2K p_\beta m_{\beta 0} \tau$ and $v_2 = K p_\beta \mathcal{G} m_{\beta 0}^2 + K p_\beta \tau^2$.

C. Fourth-order contributions to the capillary-wave Hamiltonian

The fourth-order contribution to $\mathcal{H}_\beta$ has the form

$$\mathcal{H}_\beta^2 = -\frac{K}{2} \int d\boldsymbol{R} \int_0^l dz m_{1\beta} \Delta m_{\pi\beta} \ . \qquad (44)$$

Substituting $m_{1\beta}$ [Eq. (28)] into Eq. (44), one obtains

$$\mathcal{H}_\beta^2 = -\frac{K}{2} \int d\boldsymbol{R} \int_0^l dz \, b_{1\beta} \, \Delta M_{0\beta} \, \Delta m_{\pi\beta} \ , \qquad (45)$$

where $b_{1\beta}$ and $M_{0\beta}$ are given by Eqs. (30) and (33). Since Eq. (12) leads to $\left(\partial_z^2 - p_\beta^2\right) m_{1\beta} + \Delta m_{\pi\beta} = 0$ one has

$$\Delta M_{0\beta} = \frac{\Delta m_{\pi\beta}(z\,;l)}{\left(p_\beta^2 - \partial_z^2\right) \, b_{1\beta}(z\,;l)} \ . \qquad (46)$$

This leads to the following form of the fourth-order contribution to $\mathcal{H}_\beta$:

$$\mathcal{H}_\beta^2 = -\frac{K}{2} \int d\boldsymbol{R} \int_0^l dz \, \gamma_\beta(z\,;l) \, (\Delta m_{\pi\beta})^2 \ , \qquad (47)$$

where

$$\gamma_\beta(z\,;l) = \frac{b_{1\beta}(z\,;l)}{\left(p_\beta^2 - \partial_z^2\right) \, b_{1\beta}(z;\, l)} \ . \qquad (48)$$

A similar analysis for the fourth-order α-phase contribution leads to the analogous expression

$$\mathcal{H}_\alpha^2 = -\frac{K}{2} \int d\boldsymbol{R} \int_l^\infty dz \, \gamma_\alpha(z\,;l) \, (\Delta m_{\pi\alpha})^2 \ , \qquad (49)$$

with

$$\gamma_\alpha(z\,;l) = \frac{b_{1\alpha}(z;\, l)}{(p_\alpha^2 - \partial_z^2) \, b_{1\alpha}(z;\, l)} \ . \qquad (50)$$

III. CURVATURE CONTRIBUTIONS TO THE CAPILLARY-WAVE HAMILTONIAN

The aim of this section is to show that up to fourth-order terms the capillary-wave Hamiltonian is given by Eq. (6) and to derive the expressions for the coefficients $\Sigma_{\alpha\beta}^*(l)$, $c_H^*(l)$, $c_{H^2}^*(l)$, and $c_G^*(l)$. We analyze first the free interface and then the pinned one.

A. Free α-β interface

In this case one has

$$\begin{aligned} m_{\pi\beta}^0(z;\, l) &= m_{\beta 0} \left\{ 1 - \exp[p_\beta(z - l)] \right\} \ , \\ m_{\pi\alpha}^0(z;\, l) &= m_{\alpha 0} \left\{ 1 - \exp[p_\alpha(l - z)] \right\} , \end{aligned} \qquad (51)$$

and

$$\begin{aligned} M_{0\beta}^0 &= -m_{\beta 0} \exp(-p_\beta l), \\ M_{0\alpha}^0 &= -m_{\alpha 0} \exp(p_\alpha l), \end{aligned} \qquad (52)$$

$$\gamma_\beta^0(z\,;l) = \frac{l - z}{2 p_\beta} \ , \ \gamma_\alpha^0(z\,;l) = \frac{z - l}{2 p_\alpha} \ . \qquad (53)$$

Using the above equations, the fourth-order contributions are summed up to the form

$$\begin{aligned} \mathcal{H}_\beta^{0\,2} &= -\frac{K}{2} \int d\boldsymbol{R} \int_{-\infty}^l dz \, \gamma_\beta^0 \, (\Delta m_{\pi\beta}^0)^2 \\ &= -\frac{K}{2} \int d\boldsymbol{R} \int_{-\infty}^l dz \gamma_\beta^0 [(m_{\pi\beta}^{0\prime})^2 (\Delta l)^2 \\ &\quad + (m_{\pi\beta}^{0\prime\prime})^2 \, (\nabla l)^4 + 2 \, m_{\pi\beta}^{0\prime} m_{\pi\beta}^{0\prime\prime} \Delta l (\nabla l)^2] \ , \end{aligned} \qquad (54)$$

where the prime denotes the derivative with respect to the second argument. Analogously, the fourth-order α-phase contribution has the form

$$\mathcal{H}^{0\,2}_{\alpha} = -\frac{K}{2}\int d\boldsymbol{R}\int_{l}^{\infty} dz\gamma^0_{\alpha}(\Delta m^0_{\pi\alpha})^2$$
$$= -\frac{K}{2}\int d\boldsymbol{R}\int_{l}^{\infty} dz\gamma^0_{\alpha}\Big[(m^{0\prime}_{\pi\alpha})^2(\Delta l)^2 + (m^{0\prime\prime}_{\pi\alpha})^2(\nabla l)^4 + 2\,m^{0\prime}_{\pi\alpha}\,m^{0\prime\prime}_{\pi\alpha}\,\Delta l\,(\nabla l)^2\Big]\,. \quad (55)$$

After performing the z integrations in Eqs. (54) and (55) one obtains

$$\mathcal{H}^{0\,2} = \mathcal{H}^{0\,2}_{\alpha} + \mathcal{H}^{0\,2}_{\beta}$$
$$= -\frac{K}{16}\left(m^2_{\alpha 0}p_\alpha + m^2_{\beta 0}p_\beta\right)\int d\boldsymbol{R}\,(\nabla l)^4 - \frac{K}{16}\left(\frac{m^2_{\alpha 0}}{p_\alpha} + \frac{m^2_{\beta 0}}{p_\beta}\right)\int d\boldsymbol{R}\,(\Delta l)^2 + \frac{K}{8}\left(m^2_{\beta 0} - m^2_{\alpha 0}\right)\int d\boldsymbol{R}\,\Delta l(\nabla l)^2\,. \quad (56)$$

Taking into account that up to the fourth-order terms Eqs. (3) and (4) reduce to

$$\sqrt{1+(\nabla l)^2}\,H = \frac{1}{2}[\Delta l + l_{xx}l_y^2 + l_{yy}l_x^2 - 2l_xl_yl_{xy} - \Delta l(\nabla l)^2\,],$$
$$\sqrt{1+(\nabla l)^2}\,H^2 = \frac{1}{2}(\Delta l)^2, \quad (57)$$
$$\sqrt{1+(\nabla l)^2}\,G = l_{xx}l_{yy} - l^2_{xy}$$

and including the second-order contribution

$$\mathcal{H}^{0\,1} = \frac{1}{2}\sigma_{\alpha\beta}\int d\boldsymbol{R}\,(\nabla l)^2, \quad (58)$$

one finally obtains

$$\mathcal{H}^0_{\mathrm{CW}}[l] = \int d\boldsymbol{R}\,\Big\{\sigma_{\alpha\beta}\left[\sqrt{1+(\nabla l)^2} - 1\right] + \sqrt{1+(\nabla l)^2}\,\left(c^0_H\,H + c^0_{H^2}\,H^2 + c^0_G\,G\right)\Big\}\,, \quad (59)$$

where

$$\sigma_{\alpha\beta} = \frac{K}{2}\left(p_\alpha m^2_{\alpha 0} + p_\beta m^2_{\beta 0}\right),$$
$$c^0_H = \frac{K}{2}\left(m^2_{\beta 0} - m^2_{\alpha 0}\right), \quad (60)$$
$$c^0_{H^2} = -\frac{K}{4}\left(\frac{m^2_{\alpha 0}}{p_\alpha} + \frac{m^2_{\beta 0}}{p_\beta}\right)\,,$$

while the coefficient c^0_G remains arbitrary. Note that $c^0_{H^2}$ is negative [28] and that c^0_H is antisymmetric with respect to the α-β interchange. Indeed, if one decomposes the interfacial configuration l into a flat part l_π =const and the fluctuation δl around it $l = l_\pi + \delta l$ ($\int d\boldsymbol{R}\,\delta l = 0$, $\delta l \to 0$ for $R \to \infty$), then the change of sign $\delta l \to -\delta l$ results in the change of sign of the mean curvature. But the α-β interchange and the simultaneous change of the sign of δl is the symmetry operation for the free interface and thus c^0_H must be antisymmetric under the α-β interchange, as is the case.

B. Pinned α-β interface

The case of the pinned interface requires a thorough analysis of the contribution from the β phase adsorbed on the wall; the contribution from the α phase is identical to the free case considered previously. We start from the general expression for $\mathcal{H}^2_\beta$ [see Eqs. (47), (48), and (18)]

$$\mathcal{H}^2_\beta = -\frac{K}{2}\int d\boldsymbol{R}\int_{-\infty}^{l} dz\,\gamma_\beta\,(\Delta m_{\pi\beta})^2$$
$$= -\frac{K}{2}\int d\boldsymbol{R}\int_{-\infty}^{l} dz\gamma_\beta[(m'_{\pi\beta})^2(\Delta l)^2 + (m''_{\pi\beta})^2\,(\nabla l)^4 + 2\,m'_{\pi\beta}m''_{\pi\beta}\Delta l(\nabla l)^2\,]\,. \quad (61)$$

First we briefly outline our general method employed in this paper to derive the capillary-wave Hamiltonian in the Helfrich form. Although this method is simple, it requires cumbersome calculations. It consists of the consequent application of the following procedure: an expression of the type

$$\int d\boldsymbol{R}\int_0^l dz A_1(z;l)(D_{\boldsymbol{R}}A_2)(z;l),$$

where $D_{\boldsymbol{R}}$ is a differential operator in $\boldsymbol{R}$ [for example, $D_{\boldsymbol{R}} = \Delta$; see Eq. (61)], is split into the sum of two terms with the help of an arbitrary function $f(z;l)$

$$\int d\boldsymbol{R}\int_0^l A_1(z;l)(D_{\boldsymbol{R}}A_2)(z;l)$$
$$= \int d\boldsymbol{R}\int_0^l [1-f(z;l)]A_1(z;l)(D_{\boldsymbol{R}}A_2)(z;l) + \int d\boldsymbol{R}\int_0^l f(z;l)A_1(z;l)(D_{\boldsymbol{R}}A_2)(z;l)$$

and the second term is integrated by parts. This procedure is applied three times with the use of functions $\lambda(z;l)$, $\eta(z;l)$, and $\zeta(z;l)$ in order to transform Eq. (61) into a form that contains the fourth-order contributions to $\sqrt{1+(\nabla l)^2}H$. These different contributions depend on λ, η, and ζ. The remaining steps amount to exploiting the arbitrariness of the functions λ, η, and ζ and choosing them in a such way that the coefficients multiplying these different contributions are so adjusted that they sum up to $C^*_H(l)\sqrt{1+(\nabla l)^2}H$.

Before doing that it is convenient to add zero to $\mathcal{H}^2_\beta$ in Eq. (61) written as $0 = \int d\boldsymbol{R}\int_0^l dz\,\alpha(z;l)\,(l_{xx}l_{yy} - l_{xx}l_{yy})$, where $\alpha(z;l)$ is an arbitrary function. After integrating the rhs by parts one obtains

$$
\begin{aligned}
0 &= \int d\boldsymbol{R} \int_0^l dz\, \alpha(z;l)\,(l_{xx}l_{yy} \;-\; l_{xx}l_{yy}) \\
&= \int d\boldsymbol{R}\Bigg(\int_0^l dz\, \left[\alpha(z;l)(l_{xx}l_{yy} - l_{xy}^2) - \alpha''(z;l) l_x^2 l_y^2 - 3\alpha'(z;l) l_x l_y l_{xy}\right] \\
&\quad - \left\{ \alpha'(l;l) l_x^2 l_y^2 + \alpha(l;l)\left[l_x l_y l_{xy} - \frac{1}{2}\left(l_x^2 l_{yy} + l_y^2 l_{xx}\right)\right]\right\}\Bigg) .
\end{aligned} \tag{62}
$$

The integrand in the first integral on the rhs of Eq. (62) contains the fourth-order approximation of the product $\sqrt{1+(\nabla l)^2}\,G$. With the help of auxiliary functions λ and η we perform integrations by parts as mentioned above:

$$
\begin{aligned}
-K\int d\boldsymbol{R}\int_0^l dz\gamma_\beta m'_{\pi\beta} m''_{\pi\beta}\Delta l\,(\nabla l)^2 = -K\int d\boldsymbol{R}\int_0^l dz\Bigg\{ &\gamma_\beta(1-\lambda) m'_{\pi\beta} m''_{\pi\beta}\Delta l\,(\nabla l)^2 \\
&+\frac{1}{3}\left(\gamma_\beta\lambda m'_{\pi\beta} m''_{\pi\beta}\right)'(l_x^4 + l_y^4) - \gamma_\beta\lambda(1-\eta)\, m'_{\pi\beta} m''_{\pi\beta}(l_{xx}l_y^2 + l_{yy}l_x^2) \\
&+2\left(\gamma_\beta\lambda\eta m'_{\pi\beta} m''_{\pi\beta}\right)' l_x^2 l_y^2 + 4\gamma_\beta\lambda\eta m'_{\pi\beta} m''_{\pi\beta} l_x l_y l_{xy}\Bigg\} .
\end{aligned} \tag{63}
$$

[To shorten notation we skip the arguments of functions $\gamma_\beta(z;l)$, $\lambda(z;l)$, $\eta(z;l)$, and $m_{\pi\beta}(z;l)$ in all places where it does not lead to ambiguity.] Next we collect from Eqs. (62) and (63) those terms which, after proper choice of the arbitrary functions, contribute to the mean curvature H, namely,

$$
\begin{aligned}
&\int d\boldsymbol{R}(l_x^2 l_{yy} + l_y^2 l_{xx})\left[\frac{1}{2}\alpha(l;l) - K\int_0^l dz\gamma_\beta\lambda(1-\eta) m'_{\pi\beta} m''_{\pi\beta}\right] \\
&\qquad -2l_x l_y l_{xy}\left[\frac{1}{2}\alpha(l;l) + \int_0^l dz\left(\frac{3}{2}\alpha' - 2K\gamma_\beta\lambda\eta m'_{\pi\beta}\, m''_{\pi\beta}\right)\right] - \Delta l(\nabla l)^2 K\int_0^l dz\,\gamma_\beta(1-\lambda) m'_{\pi\beta} m''_{\pi\beta} .
\end{aligned} \tag{64}
$$

Expression (64) can be written in a compact form as

$$
\int d\boldsymbol{R}\, B(l)\,[2\sqrt{1+(\nabla l)^2}\, H \;-\; \Delta l\,] \tag{65}
$$

provided the following equalities hold:

$$
\begin{aligned}
B(l) &= \frac{1}{2}\,\alpha(l;l) \;-\; K\int_0^l dz\lambda(1-\eta)\gamma_\beta\, m'_{\pi\beta}\, m''_{\pi\beta}, \\
B(l) &= \frac{1}{2}\,\alpha(l;l) \;+\; \int_0^l dz\left(\frac{3}{2}\alpha' \;-2\,K\lambda\eta\gamma_\beta\, m'_{\pi\beta}\, m''_{\pi\beta}\right), \\
B(l) &= K\int_0^l dz\,(1-\lambda)\gamma_\beta\, m'_{\pi\beta}\, m''_{\pi\beta} .
\end{aligned} \tag{66}
$$

From Eqs. (66) one obtains

$$
\begin{aligned}
\alpha(l;l) &= 2K\int_0^l dz\gamma_\beta(1-\lambda\eta) m'_{\pi\beta}\, m''_{\pi\beta}, \\
\int_0^l dz\alpha' &= -\frac{2K}{3}\int_0^l dz\lambda(1-3\eta)\gamma_\beta m'_{\pi\beta}\, m''_{\pi\beta} .
\end{aligned} \tag{67}
$$

The remaining two terms in Eq. (62) are combined with the remaining terms on the rhs of Eq. (63) and with the second term on the rhs of Eq. (61). The sum of these contributions can be transformed with the help of Eq. (67) into

$$
K\int d\boldsymbol{R}(\nabla l)^4\int_0^l dz\left[\frac{1}{3}\left(\gamma_\beta\lambda m'_{\pi\beta}\, m''_{\pi\beta}\right)' \;-\; \frac{1}{2}\gamma_\beta(m''_{\pi\beta})^2\right] . \tag{68}
$$

Thus the fourth-order β-phase contribution to the capillary-wave Hamiltonian has the form

$$\mathcal{H}_\beta^2 = -\int d\boldsymbol{R}\Bigg\{(\nabla l)^4\int_0^l dz\left[-\frac{K}{3}\left(\gamma_\beta\lambda m'_{\pi\beta}m''_{\pi\beta}\right)' + \frac{K}{2}\gamma_\beta(m''_{\pi\beta})^2\right]$$
$$-K\left[2\sqrt{1+(\nabla l)^2}\,H-\Delta l\right]\int_0^l dz\gamma_\beta(1-\lambda)m'_{\pi\beta}m''_{\pi\beta} \qquad (69)$$
$$-2K\sqrt{1+(\nabla l)^2}\,H^2\int_0^l dz(m'_{\pi\beta})^2 - \sqrt{1+(\nabla l)^2}\,G\int_0^l dz\alpha\Bigg\}.$$

The rhs of Eq. (69) has the same structure as the rhs of Eq. (2), except for the presence of the term containing Δl. The functions α and λ have to be determined on the basis of Eqs. (66). Equation (69) is supplemented by the second-order contribution $\mathcal{H}_\beta^1$ [see Eq. (39)], rewritten in the form

$$\mathcal{H}_\beta^1 = -\frac{K}{2}\int d\boldsymbol{R}\int_0^l dz\Big[m_{\pi\beta}\,m'_{\pi\beta}\Delta l + m_{\pi\beta}\,m''_{\pi\beta}(\nabla l)^2\Big]. \qquad (70)$$

Equation (69) contains, as already remarked in the Introduction, Δl, which, up to second order represents the mean curvature H. We shall use this form of $\mathcal{H}_\beta^1$ when adding it to $\mathcal{H}_\beta^2$ in order to obtain the complete expression for $\mathcal{H}_\beta$ up to the fourth-order terms. But first we rewrite the expression for $\mathcal{H}_\beta^1$ [Eq. (69)] with the help of an arbitrary function $\zeta(z;l)$ (as explained at the beginning of Sec. III) and then integrate by parts, using the crossing criterion, to obtain

$$\mathcal{H}_\beta^1 = \frac{K}{2}\int d\boldsymbol{R}\int_0^l dz\Big\{m_{\pi\beta}\,m'_{\pi\beta}(\zeta-1)\Delta l + \left[(m_{\pi\beta}\,m'_{\pi\beta}\,\zeta)' - m_{\pi\beta}\,m''_{\pi\beta}\right](\nabla l)^2\Big\}. \qquad (71)$$

If the coefficients $\sigma_\beta^1(l)$ multiplying $\frac{1}{2}(\nabla l)^2$ and $\sigma_\beta^2(l)$ multiplying $-\frac{1}{8}(\nabla l)^4$ are identical, i.e., $\sigma_\beta^1(l) = \sigma_\beta^2(l) = \Sigma_\beta^*(l)$, then the sum of these two terms can be represented, up to fourth order, as $\Sigma_\beta^*(l)\left[\sqrt{1+(\nabla l)^2}-1\right]$. At the same time we choose the remaining coefficients in such a way that the terms proportional to Δl present in $\mathcal{H}_\beta^1$ and in $\mathcal{H}_\beta^2$ cancel each other and so the contribution proportional to Δl disappears from the sum $\mathcal{H}_\beta^1+\mathcal{H}_\beta^1$. These requirements lead to the equations for λ and ζ

$$(m_{\pi\beta}\,m'_{\pi\beta}\,\zeta)' - m_{\pi\beta}\,m''_{\pi\beta} = 4\left[\gamma_\beta(m''_{\pi\beta})^2 - \frac{2}{3}(\gamma_\beta\lambda m'_{\pi\beta}m''_{\pi\beta})'\right],$$
$$m_{\pi\beta}m'_{\pi\beta}(1-\zeta) = 2\,\gamma_\beta(\lambda-1)m'_{\pi\beta}m''_{\pi\beta}\,. \qquad (72)$$

[Strictly speaking in this way one obtains two integral equations; Eqs. (72) represent the stronger requirement of the equality of the integrands in these equations.] The function $\lambda(z;l)$, which solves these equations, has the form

$$\lambda(z;l) = -3+\frac{3}{2\,\gamma_\beta m'_{\pi\beta}m''_{\pi\beta}}\times\int_z^l dl'\{4\gamma_\beta(z;l')[m''_{\pi\beta}(z;l')]^2 - [m'_{\pi\beta}(z;l')]^2\}\,. \qquad (73)$$

After substituting $\lambda(z;l)$ into the expression for $\Sigma_\beta^*(l)$ one obtains

$$\Sigma_\beta^*(l) = K\int_0^l dz\left[8(\gamma_\beta m'_{\pi\beta}m''_{\pi\beta})' - 12\gamma_\beta(m''_{\pi\beta})^2 + 4(m'_{\pi\beta})^2\right]. \qquad (74)$$

Note that the expression for the l-dependent stiffness coefficient $\Sigma_\beta^*(l)$ derived above [Eq. (74)] is different from the one derived previously within the second-order calculations [32,39]

$$\Sigma_\beta(l) = K\int_0^l dz\,(m'_{\pi\beta})^2\,. \qquad (75)$$

Nonetheless, the limiting values of these two functions are the same: $\Sigma_\beta^*(\infty) = \Sigma_\beta(\infty) = \frac{K}{2}m_{\beta 0}^2 p_\beta$.

The coefficient $2B(l)$ multiplying the mean curvature in Eq. (65) is given in Eq. (66); for the time being we call this coefficient $\bar{C}_{\beta H}^*(l)$. After inserting the expression for λ [Eq. (73)] into Eq. (66) one obtains

$$\bar{C}_{\beta H}^*(l) = \int_0^l dl'\,\left[\Sigma_\beta^*(l') - \Sigma_\beta(l')\right]. \qquad (76)$$

Next we determine the coefficients $C_{\beta H^2}^*(l)$ and $C_{\beta G}^*(l)$. From Eqs. (61) and (69) one obtains

$$C_{\beta H^2}^*(l) = -2K\int_0^l dz\,\gamma_\beta(m'_{\pi\beta})^2\,, \qquad (77)$$

$$\bar{C}_{\beta G}^*(l) = \int_0^l dz\alpha(z;l)\,. \qquad (78)$$

To derive the explicit expression for $\bar{C}_{\beta G}^*(l)$ the solution of the integral equation (67) for $\alpha(z;l)$ has to be known. We proceed similarly as before and equate the integrands on both sides of this equation; the solution of such obtained equation is

$$\alpha(z;l) = \alpha(z;z) - \frac{2K}{3}\int_z^l dl'\lambda(z;l')\,[1-3\eta(z;l')] \times\gamma_\beta(z;l')m'_{\pi\beta}(z;l')m''_{\pi\beta}(z;l') \,. \quad (79)$$

It is, however, straightforward to check that Eq. (76) leads to the wrong limiting value of $\bar{C}^*_{\beta H}(l)$; the correct value is given by Eq. (60). The reason for this apparent disagreement is similar to the free case discussed in detail in Appendix B. To obtain the correct expression (for details, see Appendix D) one has to split, analogously to the free case, the expression $\int d\boldsymbol{R}\sqrt{1+(\nabla l)^2}\mathcal{C}^*_{\beta G}(l)\,G$ into two parts, one of which is transformed via integrations by parts into the expression contributing to $\int d\boldsymbol{R}\sqrt{1+(\nabla l)^2}\mathcal{C}^*_{\beta H}(l)\,H$. After including this additional contribution one obtains finally

$$\mathcal{C}^*_{\beta H}(l) = \frac{K}{2}m^2_{\beta 0} - \int_l^\infty dl'\ [\Sigma^*_\beta(l') - \Sigma_\beta(l')] \quad (80)$$

and the expression for

$$\mathcal{C}^*_{\beta G}(l) = \frac{4K}{3}\int_0^l dl'\left[\int_0^{l'} dz\gamma_\beta(z;l')\,m'_{\pi\beta}(z;l') \times m''_{\pi\beta}(z;l') + \frac{1}{8}Km^2_{\beta 0}\right] +\frac{1}{3}\int_0^l dl'\int_0^{l'} dl''\ [\Sigma^*_{\alpha\beta}(l'') - \Sigma_{\alpha\beta}(l'')] + C\,, \quad (81)$$

where C is an arbitrary constant. The α-phase contributions to the coefficients $\Sigma^*_{\alpha\beta}(l), \mathcal{C}^*_H, \mathcal{C}^*_{H^2}(l)$ are identical to the free case (studied in Sec. III A) and equal $\frac{1}{2}Kp_\alpha m^2_{\alpha 0}, -\frac{K}{2}m^2_{\alpha 0}, -\frac{Km^2_{\alpha 0}}{4p_\alpha}$, respectively. The full l-dependent coefficients are obtained as the sums of the α and β contributions.

C. Asymptotic behavior of the coefficients $\Sigma^*_{\alpha\beta}(l), \mathcal{C}^*_H(l), \mathcal{C}^*_{H^2}(l), \mathcal{C}^*_G(l)$

In previous sections we have derived the integral expressions for the l-dependent coefficients $\Sigma^*_{\alpha\beta}(l), \mathcal{C}^*_H(l), \mathcal{C}^*_{H^2}(l), \mathcal{C}^*_G(l)$ [see Eqs. (74), (80), (77), and (81)] and we checked that these expressions have the proper limiting values [see Eqs. (60)]. It is straightforward though cumbersome to derive the explicit expressions for the coefficients $\Sigma^*_{\alpha\beta}(l), \mathcal{C}^*_H(l), \mathcal{C}^*_{H^2}(l), \mathcal{C}^*_G(l)$, at least for large l. Details of the derivation of $\Sigma^*_{\alpha\beta}(l)$ are given in Appendix C. The derivation of the l dependence of the remaining coefficients is similar and thus below we present only the results. It turns out that up to the X^2 terms $[(X = \exp(-p_\beta l)]$ each of these coefficients can be written in the general form

$$\mathcal{C}(l) = \mathcal{C}_0 + (\mathcal{C}_{10} + \mathcal{C}_{11}p_\beta l)\,X + \left[\mathcal{C}_{20} + \mathcal{C}_{21}p_\beta l + \mathcal{C}_{22}(p_\beta l)^2\right] X^2 + \dots\,. \quad (82)$$

In particular,

$$\Sigma^*_{\alpha\beta}(l) = \sigma_{\alpha\beta} + \omega^*_{10}\,X + \left[\omega^*_{20} + \omega^*_{21}p_\beta l + \omega^*_{22}(p_\beta l)^2\right] X^2 + \dots, \quad (83)$$

where

$$\sigma_{\alpha\beta} = \tfrac{1}{2}K\left(m^2_{\alpha 0}p_\alpha + m^2_{\beta 0}p_\beta\right),$$
$$\omega^*_{10} = 2Kp_\beta m_{\beta 0}\tau,$$
$$\omega^*_{20} = \tfrac{1}{2}Kp_\beta m^2_{\beta 0}\left[\mathcal{G}^2 + 16\mathcal{G} - 1 - 4(\mathcal{G}+5)\rho p^2_\beta\right], \quad (84)$$
$$\omega^*_{21} = Kp_\beta m^2_{\beta 0}\left(\mathcal{G}^2 + 10\mathcal{G} - 1 + 4\rho p^2_\beta\right),$$
$$\omega^*_{22} = -4Kp_\beta m^2_{\beta 0}\mathcal{G}\,;$$

$$\mathcal{C}^*_H(l) = c^0_H + \left[q_{20} + q_{21}p_\beta l + q_{22}(p_\beta l)^2\right] X^2 + \dots\,, \quad (85)$$

where

$$c^0_H = \frac{K}{2}\left(m^2_{\beta 0} - m^2_{\alpha 0}\right),$$

$$q_{20} = -\frac{1}{4}Km^2_{\beta 0}\left[\mathcal{G}^2 + 18\mathcal{G} - 1 - 4(\mathcal{G}+4)\rho p^2_\beta\right] + 4K\tau^2,$$

$$q_{21} = -\frac{1}{2}Km^2_{\beta 0}(\mathcal{G}^2 + 8\mathcal{G} - 1 + 4\rho p^2_\beta), \quad (86)$$

$$q_{22} = 2Km^2_{\beta 0}\mathcal{G}\ ;$$

$$\mathcal{C}_{H^2}(l) = c^0_{H^2} + s_{10}X + \left[s_{20} + s_{21}(p_\beta l) + s_{22}(p_\beta l)^2\right] X^2 + \dots\,, \quad (87)$$

where

$$c^0_{H^2} = -\frac{K}{4}\left(\frac{m^2_{\beta 0}}{p_\beta} + \frac{m^2_{\alpha 0}}{p_\alpha}\right),$$

$$s_{10} = -\frac{K\tau m_{\beta 0}}{p_\beta},$$

$$s_{20} = \frac{K\tau^2}{p_\beta} - \frac{Km^2_{\beta 0}}{4p_\beta}\left[\mathcal{G}^2 - 1 + 2(2\mathcal{G}+1)\rho p^2_\beta\right], \quad (88)$$

$$s_{21} = -\frac{Km^2_\beta}{2p_\beta}\left(\mathcal{G} - 1 + 4\rho p^2_\beta\right),$$

$$s_{22} = \frac{2Km^2_{\beta 0}}{p_\beta}\mathcal{G}\ ;$$

and

$$\mathcal{C}_G^*(l) = c + r_{10}X + \left[r_{20} + r_{21}(p_\beta l) + r_{22}(p_\beta l)^2\right] X^2 + \ \dots\ , \quad (89)$$

where

$$r_{10} = \frac{K\tau m_{\beta 0}}{p_\beta},$$

$$r_{20} = \frac{K m_{\beta 0}^2}{12 p_\beta}\left[3\mathcal{G}^2 + 9\mathcal{G} - 3 - 2(3\mathcal{G}+1)\rho p_\beta^2\right] + \frac{K\tau^2}{6p_\beta},$$

$$r_{21} = \frac{K m_{\beta 0}}{4 p_\beta}\left(\mathcal{G}^2 - 1 + 4\rho p_\beta^2\right), \quad (90)$$

$$r_{22} = -\frac{2K m_{\beta 0}^2}{2 p_\beta}\mathcal{G},$$

and c is an arbitrary constant.

We have already remarked that the expression for the l-dependent surface stiffness coefficient as obtained within fourth-order theory, i.e., $\Sigma_{\alpha\beta}^*(l)$, is different from the one obtained within second-order theory, i.e., $\Sigma_{\alpha\beta}^*(l) \neq \Sigma_{\alpha\beta}(l)$. This difference is already reflected in the form of the coefficients ω_{20}^*, ω_{21}^*, and ω_{22}^* as compared with those obtained from second-order theory. In particular, $\omega_{22}^* = -4Kp_\beta m_\beta^2 \mathcal{G} < 0$ while $\omega_{22} \equiv 0$; thus the dominant next-to-leading-order correction term to $\sigma_{\alpha\beta}$ for large $p_\beta l$ is not of the form $\omega_{21}\, p_\beta l\, X^2$, as in the case of $\Sigma_{\alpha\beta}(l)$, but is of the form $\omega_{22}^*(p_\beta l)^2 X^2$. Moreover, we observe that for the coefficients $\Sigma_{\alpha\beta}^*(l), \mathcal{C}_{H^2}^*(l), \mathcal{C}_G^*(l)$ the leading-order correction term to the free case is of the form $C_{10}X$, i.e., $C_{11} = 0$ [see Eq. (82)]. In these cases $C_{10} \sim \tau$, which means that for $\tau = 0$ the leading-order correction term has the form $\left[C_{20} + C_{21}p_\beta l + C_{22}(p_\beta l)^2\right] X^2$. Note that similar property holds also in the case of the effective interface potential $V(l)$ [see Eqs. (6) and (39)–(43)]. In the case of the coefficient $\mathcal{C}_H^*(l)$ the term proportional to X is missing.

IV. CONCLUSION

Using the mean-field version of the LGW theory supplemented by the crossing criterion, we have derived the capillary-wave Hamiltonian for an interface fluctuating in the presence of a flat substrate. Our derivation includes terms up to fourth-order in the spatial derivatives; such terms contribute to the mean and to the Gaussian curvatures and also to the change of the interfacial area, i.e., to $\sqrt{1+(\nabla l)^2} - 1 \simeq \frac{1}{2}(\nabla l)^2 - \frac{1}{8}(\nabla l)^4$. The capillary-wave Hamiltonian is then unambigously written in a form analogous to the Helfrich Hamiltonian for a membrane; the difference is that, in addition to the curvature terms, it contains the standard term describing the cost in free energy due to the change of the interfacial area. All the coefficients that enter this fourth-order Hamiltonian are found to be functions of the local distance l between the fluctuating interface and the substrate. Thus the distance l enters not only into the expressions for the mean and the Gaussian curvatures and into the interfacial area change but also into the coefficients multiplying these geometrical factors in the capillary-wave Hamiltonian. We have derived the general expressions for these functions and we have checked that in the limit of the free interface they reduce to the expressions derived in an independent way. We have also explicitly determined the leading-order terms in these functions when $p_\beta l$ becomes large. In this case these functions have a characteristic structure: the sum of $\exp(-jp_\beta l)$ multiplied by the polynomials in $p_\beta l$ of order not higher than j, $j = 0, 1, \dots$. This structure has been already determined by Jin and Fisher [31] in the case of the surface stiffness function in their analysis of second-order contributions to the capillary-wave Hamiltonian. Here we showed that this structure extends also to the functions multiplying the curvature contributions. Moreover, we have indicated that the surface stiffness function as obtained within fourth-order theory, i.e., $\Sigma_{\alpha\beta}^*(l)$, differs from the expression obtained within second-order theory, i.e., $\Sigma_{\alpha\beta}(l)$. This difference in the general expressions derived for $\Sigma_{\alpha\beta}^*(l)$ and for $\Sigma_{\alpha\beta}(l)$ is reflected in the values of the coefficients multiplying $\exp(-2p_\beta l)$ in the large $p_\beta l$ expansion of $\Sigma_{\alpha\beta}^*(l)$ and $\Sigma_{\alpha\beta}(l)$. It turns out that not only $\omega_{20}^* \neq \omega_{20}$ and $\omega_{21}^* \neq \omega_{21}$, but also $\omega_{22}^* < 0$ while $\omega_{22} = 0$. This fact may have important consequences. Indeed, it was shown by Jin and Fisher [32] that the value of the coefficient ω_{21} ($\omega_{21} < 0$) is of crucial importance for the predictions of the functional renormalization-group analysis of the wetting transitions based on the second-order Hamiltonian. Since in fourth-order theory not only $\omega_{21}^* \neq \omega_{21}$ but also the term $(p_\beta l)^2 \exp(-2p_\beta l)$ is present, one can speculate that renormalization-group analysis based on the fourth-order Hamiltonian might lead to different predictions with respect to the location of the separatrix between the first- and second-order wetting transitions. This problem is left for the future studies.

Finally, let us comment on the ambiguity in the structure of the l-dependent coefficients within the second-order-theory mentioned in the Introduction and on how this ambiguity is reflected in the renormalization-group flow equations within this theory. It is straightforward to check that the functional renormalization of the second-order Hamiltonian of the form

$$\mathcal{H}_{\rm CW}[l] = \int d\boldsymbol{R}\left[\frac{1}{2}\,\Sigma_{\alpha\beta}^*(l)\,(\nabla l)^2 + \frac{1}{2}\mathcal{C}_H^*(l)\,\Delta l + V(l)\right] \quad (91)$$

leads to the flow equations

$$\frac{\partial \Sigma_{\alpha\beta}^{*t}(l)}{\partial t} = \zeta l \frac{\partial \Sigma_{\alpha\beta}^{*t}(l)}{\partial l} + \omega \xi_\beta^2 \frac{\partial^2 \Sigma_{\alpha\beta}^{*t}(l)}{\partial l^2}, \quad (92)$$

$$\frac{\partial \mathcal{C}_H^{*t}(l)}{\partial t} = -\zeta \mathcal{C}_H^{*t} + \zeta l \frac{\partial \mathcal{C}_H^{*t}(l)}{\partial l} + \omega \xi_\beta^2 \frac{\partial^2 \mathcal{C}_H^{*t}(l)}{\partial l^2}, \quad (93)$$

$$\frac{\partial V^t(l)}{\partial t} = (d-1)V^t(l) + \zeta l \frac{\partial V^t(l)}{\partial l} + \omega \xi_\beta^2 \frac{\partial^2 V^t(l)}{\partial l^2} + \omega \xi_\beta^2 \Lambda^2 \left[\Sigma^{*t}_{\alpha\beta}(l) - \frac{\partial \mathcal{C}^{*t}_H(l)}{\partial l} \right] , \quad (94)$$

where $\zeta = \frac{3-d}{2}$ (in this paper d=3), Λ is the momentum cutoff, t is the usual length rescaling parameter, and ω is the capillary parameter given by

$$\omega = \frac{k_B T_w 2^{2-d} \Lambda^{d-3}}{\pi^{\frac{d-1}{2}} \Gamma(\frac{d-1}{2}) \sigma_{\alpha\beta}} ; \quad (95)$$

for details of the renormalization procedure see [32]. To analyze the order of the wetting transition one has to know the renormalized potential $V^t(l)$. This is obtained by solving Eq. (94) with the input $\Sigma^{*t}_{\alpha\beta}(l)$ and $\mathcal{C}^{*t}_H$ derived by solving Eqs. (92) and (93). It turns out, however, that if one uses Eq. (76), then the last term on the rhs of Eq. (94) is replaced by $\omega \xi_\beta^2 \Lambda^2 \Sigma^t_{\alpha\beta}(l)$ and thus Eq. (94) takes exactly the form used by Jin and Fisher [32]. Moreover, using Eqs. (76), (92), and (93), one can show that the function $\Sigma^t_{\alpha\beta}(l)$ fulfills exactly the same equation as $\Sigma^{*t}_{\alpha\beta}(l)$ does, i.e., Eq. (92), and thus the solution of the problem is identical to the one given in [32].

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APPENDIX A

In this appendix we show that the β-phase contribution to the capillary-wave Hamiltonian is given by Eq. (39). For this purpose, into each of the expressions for $\mathcal{H}_{\beta i}$, $i = 1, \ldots, 4$, we insert Eq. (26) and perform integration by parts, taking into account the crossing criterion Eq. (29). In this way one obtains the following expressions for the $2n$th-order contributions $\mathcal{H}^n_{\beta i}$, $n \geq 1$:

$$\mathcal{H}^n_{\beta 1} = -K \int d\boldsymbol{R} \left\{ m_n m'_{\pi\beta}|_{z=0} + \frac{1}{2} \sum_{i+j=n} m_i \, m'_j|_{z=0} + \int_0^l dz \left[m_n m''_{\pi\beta} + \frac{1}{2} \sum_{i+j=n} m_i \, m''_j \right] \right\} , \quad (A1)$$

$$\mathcal{H}^n_{\beta 2} = -K \int d\boldsymbol{R} \int_0^l dz \left\{ m_{n-1} \Delta m_{\pi\beta} + \frac{1}{2} \sum_{i+j=n-1} m_i \, \Delta m_j \right\} , \quad (A2)$$

$$\mathcal{H}^n_{\beta 3} = K p_\beta^2 \int d\boldsymbol{R} \int_0^l dz \left\{ m_n \, (m_{\pi\beta} - m_{\beta 0}) + \frac{1}{2} \sum_{i+j=n} m_i \, m_j \right\} , \quad (A3)$$

$$\mathcal{H}^n_{\beta 4} = \int d\boldsymbol{R} \left\{ -h_1 m_n|_{z=0} - g m_n m_{\pi\beta}|_{z=0} - \frac{g}{2} \sum_{i+j=n} m_i m_j|_{z=0} \right\} . \quad (A4)$$

After summing the above contributions [Eqs. (A1)–(A4)], one obtains

$$\mathcal{H}^n_\beta = -\int d\boldsymbol{R} \left[\left(K m'_{\pi\beta} + h_1 + g m_{\pi\beta} \right) m_n \Big|_{z=0} - \frac{1}{2} \sum_{i+j=n} \left(K m'_j + g m_j \right) m_i \Big|_{z=0} \right]$$

$$- K \int d\boldsymbol{R} \int_0^l dz \left\{ m_n \left[m''_{\pi\beta} - p_\beta^2 (m_{\pi\beta} - m_{\beta 0}) \right] + \frac{1}{2} \sum_{i+j=n} (p_\beta^2 m_j - m''_j) m_i \right\} \quad (A5)$$

$$-\frac{K}{2} \int d\boldsymbol{R} \int_0^l dz \left[\sum_{i+j=n-1} m_i \Delta m_j + 2 m_{n-1} \Delta m_{\pi\beta} \right] .$$

The first two terms vanish due to the boundary condition at $z = 0$ [Eq. (13)] and the third term vanishes because $m_{\pi\beta}$ fulfills Eq. (12). Finally, one obtains

$$\sum_{n=2}^{\infty} \mathcal{H}_\beta^n = \frac{K}{2} \sum_{n=2}^{\infty} \int d\boldsymbol{R} \int_0^l dz \left[\sum_{i+j=n} m_i \Delta m_{j-1} - \sum_{i+j=n-1} m_i \Delta m_j - 2 m_{n-1} \Delta m_{\pi\beta} \right]. \quad \text{(A6)}$$

It is straightforward to check that

$$\sum_{n=2}^{\infty} \left(\sum_{i+j=n} m_i \Delta m_{j-1} - \sum_{i+j=n-1} m_i \Delta m_j \right) = \sum_{n=2}^{\infty} m_{n-1} \Delta m_{\pi\beta}, \quad \text{(A7)}$$

so that

$$\sum_{n=2}^{\infty} \mathcal{H}_\beta^n = -\frac{K}{2} \sum_{n=1}^{\infty} \int d\boldsymbol{R} \int_0^l dz\, m_n \Delta m_{\pi\beta} . \quad \text{(A8)}$$

After adding the lowest-order contribution $\mathcal{H}_\beta^1$ derived previously

$$\mathcal{H}_\beta^1 = -\frac{K}{2} \int d\boldsymbol{R} \left[\int_0^l dz m_{\pi\beta} \Delta m_{\pi\beta} + V(l) \right], \quad \text{(A9)}$$

one finally obtains

$$\mathcal{H}_\beta = \int d\boldsymbol{R} \left[-\frac{K}{2} \int_0^l dz\, m_{\pi\beta}\, \Delta\, m_\beta + V(l) \right] . \quad \text{(A10)}$$

APPENDIX B

In this appendix we analyze the asymptotic behavior of various coefficients present in Eq. (6) in the limit of *free* interface, i.e., for $l \to \infty$. For the free case one can use the results derived for the pinned interface in which the expressions for $m_{\pi\gamma}^0$ and γ_γ^0 are given by Eqs. (51) and (53) and the z integration extends from $-\infty$ to l. A straightforward calculation shows that in the free case

$$\lambda^0(z;l) = 0,$$
$$\alpha^0(z;\, l) = \alpha^0(z;\, z) = -\frac{K}{4} m_{\beta 0}^2, \quad \text{(B1)}$$
$$\bar{c}_{\beta\, H}^0 = -\frac{1}{4} K\, m_{\beta 0}^2 .$$

$\bar{c}_{\beta\, H}^0$ is different from the one obtained within the direct analysis of the free case [see Sec. III A, Eq. (60)]. At the same time the Gaussian curvature contribution $\int d\boldsymbol{R} \int_{-\infty}^l dz \sqrt{1 + (\nabla l)^2}\, G$ contains the divergent integral over z, which, however, is multiplied by the vanishing integral $\int d\boldsymbol{R} \sqrt{1 + (\nabla l)^2}\, G$. Below we point to the fact that this divergence is in fact apparent; we also find the missing part $\delta c_{\beta H}^0$ of $c_{\beta\, H}^0$ leading to agreement with the previously found expression Eq. (60). That part of $\mathcal{H}_\beta$ that contains the Gaussian curvature is rewritten as

$$\int d\boldsymbol{R} \int_{-L}^l dz \sqrt{1 + (\nabla l)^2} G = \int d\boldsymbol{R}\, [(l - a) + (a - L)]\, \sqrt{1 + (\nabla l)^2}\, G = \int d\boldsymbol{R} l \sqrt{1 + (\nabla l)^2}\, G - a \int d\boldsymbol{R} \sqrt{1 + (\nabla l)^2}\, G, \quad \text{(B2)}$$

where a is an arbitrary constant. Integration by parts gives

$$\int d\boldsymbol{R} l \sqrt{1 + (\nabla l)^2}\, G = -3 \int d\boldsymbol{R} \sqrt{1 + (\nabla l)^2}\, H . \quad \text{(B3)}$$

Equation (B2) holds for a free system with a finite extension along the z axis and L represents the cutoff; we employ it also in the limit $L \to \infty$. Finally,

$$-\frac{K}{4} m_{\beta 0}^2 \int d\boldsymbol{R} \int_{-\infty}^l dz \sqrt{1 + (\nabla l)^2}\, G = \frac{3K}{4} m_{\beta 0}^2 \int d\boldsymbol{R} \sqrt{1 + (\nabla l)^2}\, H + A_0 \int d\boldsymbol{R} \sqrt{1 + (\nabla l)^2}\, G\, , \quad \text{(B4)}$$

where A_0 is an arbitrary constant. We see that the parameter $c_{\beta G}^0$ multiplying the Gaussian curvature remains arbitrary [34] and that both contributions to $c_{\beta H}^0$, i.e., $\bar{c}_{\beta H}^0$ in Eq. (B1) and the contribution contained in Eq. (B4), add up to the correct form of the $c_{\beta H}^0$ coefficient as given by Eq. (60).

APPENDIX C

In this appendix we derive the explicit expression for the function $\Sigma_\beta^*(l)$ up to the terms proportional to X^2. The expression for $\Sigma_\beta^*(l)$ [Eq. (74)] depends not only on $m_{\pi\beta}$ [Eqs. (16) and (17)], but also on the function $\gamma_\beta(z;l)$ [Eqs. (48) and (18)]. $\gamma_\beta(z;l)$ can be written in the form

$$\gamma_\beta(z;l) = [A + Bz + (C + Dz) \exp(-2p_\beta z)] \times [1 - \mathcal{G} \exp(-2p_\beta z)]^{-1}, \quad \text{(C1)}$$

where

$$A = \frac{l}{2p_\beta} + \frac{\mathcal{G}l - p_\beta\rho}{p_\beta(1-\mathcal{G}X^2)}, \quad B = -\frac{1}{2p_\beta},$$
$$C = -\frac{\mathcal{G}l}{2p_\beta} + \frac{\mathcal{G}^2X^2l - p_\beta\rho}{p_\beta(1-\mathcal{G}X^2)}, \quad D = -\frac{\mathcal{G}}{2p_\beta}. \tag{C2}$$

To derive the explicit expressions for the coefficients multiplying the powers of X in $\Sigma^*_{\alpha\beta}(l)$ one has to evaluate the integrals

$$K_j = \int_0^l dz \exp(-p_\beta z)\frac{\exp(jp_\beta z)}{1-\mathcal{G}\exp(-2p_\beta z)}, \tag{C3}$$

$$L_j = \int_0^l dz \exp(-p_\beta z)\frac{z\exp(jp_\beta z)}{1-\mathcal{G}\exp(-2p_\beta z)} \tag{C4}$$

for $j=-3,-1,1,3$. While the integrals K_j are elementary, the L_j's are not. They can be expressed in terms of the dilogarithmic function $Li_2(t)$ [52]

$$Li_2(t) = -\int_0^t ds\frac{\ln(1-s)}{s}, \tag{C5}$$

which has the following power series expansion for $t^2 \le 1$:

$$Li_2(t) = \sum_{n=1}^{\infty}\frac{t^n}{n^2}. \tag{C6}$$

Straightforward and tedious calculations, which use expansion (C6) and the identity [52]

$$Li_2(t) + Li_2(1-t) = \frac{\pi^2}{6} - \ln(t)\ln(1-t), \tag{C7}$$

lead to the results

$$\Sigma^*_{\alpha\beta}(l) = \sigma_{\alpha\beta} + \omega^*_{10}X + [w^*_{20} + \omega^*_{21}p_\beta l + \omega^*_{22}(p_\beta l)^2]X^2 + \dots, \tag{C8}$$

where

$$\sigma_{\alpha\beta} = \frac{1}{2}K(p_\alpha m^2_{\alpha 0} + p_\beta m^2_{\beta 0}),$$
$$\omega^*_{10} = 2Kp_\beta m_{\beta 0}\tau,$$
$$\omega^*_{20} = \frac{1}{2}Kp_\beta m^2_{\beta 0}\left[\mathcal{G}^2 + 16\mathcal{G} - 4(\mathcal{G}+5)\rho p^2_\beta\right], \tag{C9}$$
$$\omega^*_{21} = Kp_\beta m^2_{\beta 0}\left[\mathcal{G}^2 + 10\mathcal{G} - 1 + 4\rho p^2_\beta\right],$$
$$\omega^*_{22} = -4Kp_\beta m^2_{\beta 0}\mathcal{G}.$$

Note that $\Sigma^*_{\alpha\beta}(l)$ [Eq. (C7)] has the same structure and the same limiting value $\Sigma^*_{\alpha\beta}(\infty) = \sigma_{\alpha\beta}$ as the function $\Sigma_{\alpha\beta}(l)$ proposed by Jin and Fisher [31] within the second-order analysis. Moreover, $\omega^*_{10} = \omega_{10}$ while the coefficients ω^*_{20}, ω^*_{21}, and ω^*_{22} are different from those obtained in [31].

APPENDIX D

In this appendix we analyze the Gaussian curvature contribution $\int d\boldsymbol{R}\sqrt{1+(\nabla l)^2}\,C^*_{\beta G}(l)\,G$ to the capillary-wave Hamiltonian. We want to check if also in the pinned case, similarly to the free case studied in Appendix B, one can extract from this term (via integration by parts) a contribution to the mean curvature term, which after combining with the already existing contribution $\bar{C}^*_{\beta H}(l)$ [Eq. (76)] gives the correct limiting value of the coefficient $C^*_{\beta H}(l)$. For this purpose it is convenient to rewrite the expression for $\bar{C}^*_{\beta G}(l)$ [Eq. (78)] as

$$\bar{C}^*_{\beta G}(l) = \int_0^l dz\alpha(z;l) = 2K\int_0^l dz\int_0^z dz'\left\{\gamma_\beta(z';z)\left[1-\eta(z';z)\lambda(z';z)\right]m'_{\pi\beta}(z';z)m''_{\pi\beta}(z';z)\right\}$$
$$-\frac{2K}{3}\int_0^l dz\int_z^l dl'\left\{\gamma_\beta(z;l')\lambda(z;l')\left[1-3\eta(z;l')\right]m'_{\pi\beta}(z;l')m''_{\pi\beta}(z;l')\right\}. \tag{D1}$$

Changing the order of integration in the second term on rhs of Eq. (D1) leads to

$$\bar{C}^*_{\beta G}(l) = 2K\int_0^l dl'\int_0^{l'} dz\left\{\gamma_\beta(z;l')\left[1-\frac{1}{3}\lambda(z;l')\right]\times m'_{\pi\beta}(z;l')m''_{\pi\beta}(z;l')\right\}. \tag{D2}$$

Note that the above expression is η independent. After using the expression for λ [Eq. (73)] and changing the order of integration one obtains

$$\bar{C}^*_{\beta G}(l) = \frac{4K}{3}\int_0^l dl'\int_0^{l'} dz\,\gamma(z;l')m'_{\pi\beta}(z;l')m''_{\pi\beta}(z;l')$$
$$+\frac{1}{3}\int_0^l dl'\int_0^{l'} dl''\left[\Sigma^*_\beta(l'') - \Sigma_\beta(l'')\right]. \tag{D3}$$

It is convenient to use the notation

$$K\int_0^l dz\gamma m'_{\pi\beta}m''_{\pi\beta} = A_0 + A_1(l) \tag{D4}$$

and

$$\int_0^l dl' \left[\Sigma^*_\beta(l') - \Sigma_\beta(l')\right] = B_0 + B_1(l) \,, \qquad \text{(D5)}$$

where A_0, B_0 are constants and $A_1(l), B_1(l)$ decay exponentially to zero for $l \to \infty$. Using the above defined functions and the identity (up to fourth-order terms)

$$\int d\boldsymbol{R}\sqrt{1+(\nabla l)^2}\, l\, G = -3 \int d\boldsymbol{R}\sqrt{1+(\nabla l)^2}\, H \qquad \text{(D6)}$$

(see Appendix B) one obtains

$$\mathcal{C}^*_{\beta G}(l) = \frac{1}{3}\int_0^l dl' \left[4A_1(l') + B_1(l')\right] \qquad \text{(D7)}$$

and

$$\delta\mathcal{C}^*_{\beta H}(l) = -4A_0 - B_0 \,. \qquad \text{(D8)}$$

Note that because $B_1(l)$ vanishes at infinity one has

$$B_1(l) = -\int_l^\infty dl' \left[\Sigma^*_\beta(l') - \Sigma_\beta(l')\right] \,. \qquad \text{(D9)}$$

According to Eq. (D4), A_0 represents the l-independent contribution from the integral $\int_0^l dz\gamma m'_{\pi\beta} m''_{\pi\beta}$. Using the explicit expressions for $m_{\pi\beta}$ and γ_β leads to the conclusion that

$$A_0 = \lim_{l\to\infty} \left[(B_+X)'(B_+X)''(K_3A + L_3B)\right] \,, \qquad \text{(D10)}$$

where the symbols K_3, L_3, B_+, A, and B are defined in the main text and in Appendix C. A straightforward calculation gives

$$A_0 = -\frac{1}{8} m^2_{\beta 0} K \qquad \text{(D11)}$$

and finally one obtains

$$\mathcal{C}^*_{\beta H}(l) = \frac{1}{2} m^2_{\beta 0} K - \int_l^\infty dl' \left[\Sigma^*_\beta(l') - \Sigma_\beta(l)\right] \,. \qquad \text{(D12)}$$

The above expression has the correct asymptotic limit, i.e., $\frac{1}{2} m^2_{\beta 0} K$, for $l \to \infty$.

In the same way one can check that the function $\mathcal{C}^*_{\beta H^2}(l)$ [Eq. (74)] has the correct asymptotic value $-\frac{K m^2_{\beta 0}}{4p_\beta}$ [see Eq. (60)]. The final form of the coefficient $\mathcal{C}^*_{\beta G}(l)$ is

$$\mathcal{C}^*_{\beta G}(l) = \frac{4K}{3}\int_0^l dl' \left[\int_0^{l'} dz\gamma(z;l') \times m'_{\pi\beta}(z;l') m''_{\pi\beta}(z;l') + \frac{1}{8} K m^2_{\beta 0}\right] + \frac{1}{3}\int_l^\infty dl' \int_{l'}^\infty dl'' \left[\Sigma^*_\beta(l'') - \Sigma_\beta(l'')\right] + C. \qquad \text{(D13)}$$

Because of the vanishing of the integral $\int d\boldsymbol{R}\sqrt{1+(\nabla l)^2}\, G$ one can add an arbitrary constant C to the coefficient $\mathcal{C}^*_{\beta G}(l)$.

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