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From density to interface fluctuations: The origin of wavelength dependence in surface tension

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The height-height correlation function for a fluctuating interface between two coexisting bulk phases is derived by means of general equilibrium properties of the corresponding density-density correlation function. A wavelength-dependent surface tension $\gamma(\mathbf{q})$ can be defined and expressed in terms of the direct correlation function $c(\mathbf{r},\mathbf{r}')$, the equilibrium density profile $\rho_0(\mathbf{r})$, and an operator which relates density to surface configurations. Neither the concept of an effective interface Hamiltonian nor the difference in pressure is needed to determine the general structure of the height-height correlations or $\gamma(\mathbf{q})$, respectively. This result generalizes the Mecke-Dietrich surface tension $\gamma_{\mathrm{MD}}(q)$ [Phys. Rev. E **59**, 6766 (1999)] and modifies recently published criticism concerning $\gamma_{\mathrm{MD}}(q)$ [Tarazona, Checa, and Chacón, Phys. Rev. Lett. **99**, 196101 (2007)].

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Thermally excited capillary waves on the fluid interface between two phases have drawn the attention of many theoretical [1–6] and experimental [8–16] physicists for more than 100 years. Two approaches have been developed in the last decades in order to resolve the structural properties of the transition region and its thermally driven fluctuations: Density functional theory (DFT) is applied successfully in order to describe the dependence of interfacial properties on, e.g., temperature or intermolecular forces, respectively (see, e.g., [17]). In particular, the spatial dependence of the one-particle equilibrium density $\rho_0(\mathbf{r})$, which exhibits a smooth transition between two coexisting phases, can be calculated within a DFT framework.

On the other hand, capillary-wave theory (CWT), see Ref. [3], analyzes the fluctuations of a infinitely thin and planar liquid-vapor interface parallel to the xy plane $\mathbb{R}^2$. Local deviations from the planar surface configuration are represented by a random field $u(\mathbf{R})$, with $\mathbf{R}=(x,y)\in\mathbb{R}^2$, which is considered to be statistically independent of the particle distribution. Consequently, the height-height correlation function $\langle u(\mathbf{R})u(\mathbf{R}')\rangle_{\mathrm{CWT}}$ depends parametrically on the surface tension γ but does not include any particle interactions explicitly.

In 1999, Mecke and Dietrich suggested a combination of DFT and CWT. They used an isodensity criterion, i.e., $\rho(\mathbf{R},z=u(\mathbf{R}))=\mathrm{const}$, in order to define the interface $u(\mathbf{R})$ implicitly [6]. By means of a grand canonical density functional $\Omega[\rho]$, they define an effective interface Hamiltonian $\mathcal{H}[u]:=\Omega[\rho_u]-\Omega[\rho_0]$ as the difference between two free energies, associated to two *equilibrium* surface states. $\Omega[\rho_0]$ is the free energy of a planar interface parallel to the xy plane, while $\Omega[\rho_u]$ is associated to a fixed but spatially varying surface $u(\mathbf{R})$. An expansion of $\Omega[\rho_u]$ near $\Omega[\rho_0]$ [or $\rho_u(\mathbf{r})$ at $\rho_0(\mathbf{r})$, respectively] allows for an explicit expression of $\mathcal{H}[u]$ in terms of $u(\mathbf{R})$. By taking curvature corrections of the fluctuating interface into account, the resulting height correlation function is governed by a wavelength dependent surface tension $\gamma_{\mathrm{MD}}(q)$. While the included van der Waals attractions induce a decrease of $\gamma_{\mathrm{MD}}(q)$, its increase at large values of q is considered as an indication for a bending rigidity for liquid surfaces. $\gamma_{\mathrm{MD}}(q)$ has been confirmed in x-ray studies for several liquids with different accuracy [9–14].

Although the combination of DFT and CWT has been generalized to binary mixtures [7], the results depend on the particular choice of the density functional $\Omega[\rho]$ and on the validity of the curvature corrections. There exist also difficulties in analyzing the experimental scattering data in order to obtain $\gamma_{\mathrm{MD}}(q)$ [16]. In addition, recent numerical studies even cast doubt on $\gamma_{\mathrm{MD}}(q)$ [18,19]. In Ref. [19] the authors argue that $\gamma_{\mathrm{MD}}(q)$ must decrease for large values of q if derived from a DFT as in Ref. [6]. Thus alternative theoretical methods are mandatory in order to elucidate the relation between density correlations and interface correlations.

Here the height-height correlation function for a fluctuating interface between two coexisting phases is derived and expressed by means of general equilibrium properties of the corresponding density-density correlations. In principle, this approach is neither restricted to planar systems nor to liquid-vapor interfaces. An explicit expression for a wavelength-dependent surface tension $\gamma(\mathbf{q})$ can be identified, which depends on the direct correlation function $c(\mathbf{r},\mathbf{r}')$ (which is defined via the nonideal gas contributions of the *inverse* density covariance function), the equilibrium density profile $\rho_0(\mathbf{r})$, and an operator relating density and surface configurations. The expression for $\gamma(\mathbf{q})$ generalizes in particular $\gamma_{\mathrm{MD}}(q)$ and its derivation does not rely on a free energy expansion, which is one of the main criticism of Ref. [19].

We consider an equilibrium state within the grand canonical ensemble. Ensemble averages are denoted as $\langle\cdots\rangle_0$. Using the local microscopic density $\rho(\mathbf{r}):=\sum_{i=1}^{N}\delta(\mathbf{r}-\mathbf{r}_i)$ we write $\rho_0(\mathbf{r}):=\langle\rho(\mathbf{r})\rangle_0$ for the spatially dependent one-particle equilibrium density. We assume that the system exhibits a single interface between two coexisting phases. Its mathematical specification is given below. Density fluctuations are characterized by the density covariance function $G(\mathbf{r},\mathbf{r}'):=\langle[\rho(\mathbf{r})-\rho_0(\mathbf{r})][\rho(\mathbf{r}')-\rho_0(\mathbf{r}')]\rangle_0$. Since fluctuations of the interface are not independent of density fluctuations, the height-height correlation function must be related to $G(\mathbf{r},\mathbf{r}')$. In the following, we study this relationship.

It follows from probability theory that a random field $X(\mathbf{r})\in\mathbb{R}$ exists, which exhibits the same mean value and correlations as $\rho(\mathbf{r})$, i.e., $\overline{X(\mathbf{r})}=\rho_0(\mathbf{r})$ and $\overline{[X(\mathbf{r})-\rho_0(\mathbf{r})][X(\mathbf{r}')-\rho_0(\mathbf{r}')]}=G(\mathbf{r},\mathbf{r}')$, where the average $\overline{\cdots}$ is taken with respect to a suitable probability measure for X [20]. In general, the system under consideration is inhomogeneous due to the presence of an interface and thus $G(\mathbf{r},\mathbf{r}')$ does *not* depend only on $\mathbf{r}-\mathbf{r}'$. Since the first and the second moment of $X(\mathbf{r})$ are given by $\rho_0(\mathbf{r})$ and the $G(\mathbf{r},\mathbf{r}')$, $X(\mathbf{r})$ can

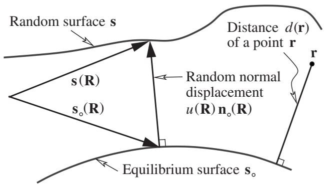

FIG. 1. The random surface $\mathbf{s}$ is generated by random displacements $u(\mathbf{R})$ of $\mathbf{s}_0(\mathbf{R})$ along its normal vector $\mathbf{n}_0(\mathbf{R})$ [see Eq. (1)]. For simplicity, the surface patch $\mathbf{s}_0$ is drawn spherically. For each spatial point $\mathbf{r}$ we write $d \equiv d(\mathbf{r})$ for its (normal) distance to the equilibrium interface $\mathbf{s}_0$ [see Eq. (3)]. Random deviations of the equilibrium density $\rho_0(\mathbf{r})$ are caused by the random displacements $u(\mathbf{R})$ of the interface as well as by a local change in volume [see Eqs. (4) and (6)].

be considered as, e.g., an inhomogeneous Gaussian process. By construction, the local microscopic density $\rho(\mathbf{r})$ and the random field $X(\mathbf{r})$ can be distinguished only by measurements or calculations of their higher correlation functions. In particular, the structure factor is not sufficient to determine the difference between $X(\mathbf{r})$ and $\rho(\mathbf{r})$. Thus $X(\mathbf{r})$ is *not* an equilibrium density but mimics the first and the second equilibrium moment of $\rho(\mathbf{r})$. We call $X(\mathbf{r})$ a random density. If, in addition, $X(\mathbf{r})$ is generated by another random field which mimics the interface configurations, the correlations of this underlying process are related to $G(\mathbf{r},\mathbf{r}')$. In the following, this procedure and the relation between the different correlation functions is discussed.

Although we will focus later on planar interfaces, we use first a slightly more abstract notation in order to isolate the core definitions and ideas from additional assumptions about the system like its symmetry.

Corresponding to the equilibrium density $\rho_0(\mathbf{r})$ we introduce an equilibrium phase boundary $\mathbf{s}_0(\mathbf{R}) \in \mathbb{R}^3$, which depends on two parameters $\mathbf{R} \in M \subseteq \mathbb{R}^2$ for a suitable subset M of $\mathbb{R}^2$. The interface $\mathbf{s}_0(\mathbf{R})$ is considered to be an isodensity contour of $\rho_0(\mathbf{r})$, i.e., $\rho_0(\mathbf{s}_0(\mathbf{R}))=\rho_c=\text{const}$ for all $\mathbf{R} \in M$. Introducing the random field $u(\mathbf{R}) \in \mathbb{R}$, a fluctuating interface $\mathbf{s}(\mathbf{R}) \in \mathbb{R}^3$ is considered to be a random field which results from $\mathbf{s}_0(\mathbf{R})$ by local normal displacements (see Fig. 1),

$$\mathbf{s}(\mathbf{R}) = \mathbf{s}_0(\mathbf{R}) + u(\mathbf{R})\mathbf{n}_0(\mathbf{R}), \tag{1}$$

where $\mathbf{n}_0(\mathbf{R})$ is the unit normal vector of the surface $\mathbf{s}_0(\mathbf{R})$. For instance, we expect $\mathbf{n}_0(x,y)=\mathbf{e}_z$ for a liquid-vapor interface in a homogeneous field acting along the z axis $\mathbf{e}_z$, while for a spherical droplet it is $\mathbf{n}_0(\vartheta,\varphi)=\mathbf{e}_r(\vartheta,\varphi)$, where $\mathbf{e}_r$ is the radial unit vector.

A relation between the random density $X(\mathbf{r})$ and the random surface $\mathbf{s}(\mathbf{R})$ establishes a relation between the density correlations $G(\mathbf{r},\mathbf{r}')$ and the correlations of $u(\mathbf{R})$. For this purpose, we require the following conditions:

$$X(\mathbf{s}(\mathbf{R})) = \rho_0(\mathbf{s}_0(\mathbf{R})) = \rho_c, \tag{2a}$$

$$\mathbf{s} \equiv \mathbf{s}_0 \Rightarrow X(\mathbf{r}) = \rho_0(\mathbf{r}) \quad \text{for all } \mathbf{r}, \tag{2b}$$

where $\mathbf{s} \equiv \mathbf{s}_0$ means the identity for all $\mathbf{R} \in M$. The condition equation (2a) generalizes the concept of an isodensity contour to $X(\mathbf{r})$. The condition equation (2b) states that $X(\mathbf{r})$ assumes its mean value if $\mathbf{s}$ is identical to $\mathbf{s}_0$. Thus we consider those $X(\mathbf{r})$ which result from $\rho_0(\mathbf{r})$ due to a change in the isodensity surface.

The above-mentioned conditions are not sufficient to determine a unique relation between $X(\mathbf{r})$ and $\mathbf{s}(\mathbf{R})$; but here we show in which way different realizations of Eqs. (2) lead to different height correlations for $u(\mathbf{R})$ exploiting only the abovementioned condition that the autocorrelation of $X(\mathbf{r}) - \rho_0(\mathbf{r})$ is given by $G(\mathbf{r},\mathbf{r}')$.

We assume that each point $\mathbf{r}$ can be represented unambiguously within the normal coordinate system of the equilibrium surface $\mathbf{s}_0$, i.e., there exist $d(\mathbf{r}) \equiv d \in \mathbb{R}$ and $\mathbf{R}(\mathbf{r}) \equiv \mathbf{R} \in M$ so that (see Fig. 1) [21]

$$\mathbf{r} = \mathbf{s}_0(\mathbf{R}) + d\mathbf{n}_0(\mathbf{R}). \tag{3}$$

A simple choice for $X(\mathbf{r})$ which fulfills Eqs. (2) is

$$X(\mathbf{r}) = \rho_0(\mathbf{r} + \mathbf{s}_0(\mathbf{R}) - \mathbf{s}(\mathbf{R})), \tag{4}$$

where $\mathbf{R} \equiv \mathbf{R}(\mathbf{r})$ [see Eq. (3)] [22]. By Eq. (1), $X(\mathbf{r})$ is generated by random displacements of the equilibrium density $\rho_0(\mathbf{r})$ along the normal vector $\mathbf{n}_0$ in whose direction $\mathbf{r}$ lies. It is worth noting that Eq. (5) is *not* a result but rather a choice for the parametrization of $X(\mathbf{r})$ based on Eqs. (2). Although Eq. (4) appears as the standard approach in order to describe capillary wave fluctuations (of planar interfaces), the specification of the conditions equations (2) is an attempt to justify this approach on a more profound principle. In particular, it does not rely on the thermodynamic conditions as, e.g., the Gibbs dividing surface concept. Here, we are interested in the generalization of Eq. (4) which is consistent with that principle.

In general, a change in density at point $\mathbf{r}$ is obtained due to a change of the infinitesimally small volume which contains the point $\mathbf{r}$. This change of the local volume at point $\mathbf{r}$ might occur from a change of the equilibrium interface even if the corresponding point on $\mathbf{s}_0$ is not shifted along $\mathbf{n}_0$. For instance, a change from a spherical droplet to an ellipsoid changes the local volume at a point $\mathbf{r}$ without moving the corresponding point on $\mathbf{s}_0$. Thus a change in curvature is one of these mechanisms. Obviously, such changes in local volume depend on the distance d between $\mathbf{r}$ and $\mathbf{s}_0$ [see Eq. (3) and Fig. 1]. Due to the isodensity condition this effect vanishes for $d=u(\mathbf{R})$, i.e., if $\mathbf{r}$ lies on $\mathbf{s}(\mathbf{R})$. Equally, if the amplitude of the interface fluctuations are sufficiently small, the local volume within the bulk phases does not change. Thus this effect vanishes for $d \gg \xi$, where ξ is the effective width of the interface, i.e., the width of the transition region in which $\rho_0(\mathbf{r})$ differs considerably from the bulk densities. For steplike profiles it is $\xi=0$. In such cases we do not expect such a change in local volume due to other mechanisms. A change in local volume that affects the local density $\rho_0(\mathbf{r})$ can be associated with a local compressibility. Therefore the following generalizations of Eq. (4) are intended to take into account the local compressibility.

The incompleteness of the standard capillary wave ansatz Eq. (4) can be understood equally from the physical picture that density fluctuations are present in each spatial direction and not only along the normal direction of the interface. This lack in description could be adjusted by taking tangential fluctuations (parallel to the interface) into account. On the other hand, tangential density fluctuations affect the normal density fluctuations due to the isodensity condition of the interface. Therefore we expect an additional term in Eq. (4) which projects tangential density fluctuation on normal density fluctuations, similar as it is done in the projector operator formalism. This projector includes the ratio of correlation functions associated with tangential and normal fluctuations, respectively. Furthermore, it depends in general on the distance to the interface and on the wavelength of the fluctuation: For large distances the tangential density fluctuations are not expected to affect the normal density fluctuation which implies that the projected contribution disappears independently of the wavelength of the fluctuation. Close to the interface, a long-wavelength tangential density fluctuation will barely have an influence on the local interfacial structure due to the isodensity condition, i.e., such tangential fluctuations shall be rather aligned by the global structure of the interface. Thus the projector is expected to vanish in that case. For the same reason, the local interfacial structure depends on short-wavelength tangential fluctuation close to the interface. For instance, the interface might bend locally due to a tangential density fluctuation above or below the interface. At this point the interpretation in terms of tangential density fluctuations becomes similar to the above-mentioned local compressibility picture. Therefore the interfacial width ξ can be considered also as the range of the coupling between these two types of fluctuations.

After these pictorial remarks we introduce the $\mathbf{r}$-dependent linear operator $\mathsf{L}(\mathbf{r})$ which acts on the surface $\mathbf{s}(\mathbf{R})$ [and $\mathbf{s}_0(\mathbf{R})$] via

$$\mathsf{L}(\mathbf{r})\mathbf{s}(\mathbf{R}) := \int_M \mathbf{L}(\mathbf{r};\mathbf{R},\mathbf{R}')\mathbf{s}(\mathbf{R}')d\mathbf{R}', \tag{5}$$

with a 3×3-matrix $\mathbf{L}(\mathbf{r};\mathbf{R},\mathbf{R}')$ as the integral kernel. Since $\mathsf{L}(\mathbf{r})$ is intended to model the effect of local compressibility we expect $\mathsf{L}(\mathbf{r})\mathbf{s}_0(\mathbf{R})=\mathbf{0}$ for all $\mathbf{R}\in M$ and all $\mathbf{r}$ due to the isodensity condition. Thus it is $\mathsf{L}(\mathbf{r})\mathbf{s}(\mathbf{R})=\mathsf{L}(\mathbf{r})(u(\mathbf{R})\mathbf{n}_0(\mathbf{R}))$. Equally we assume $\mathsf{L}(\mathbf{0})=\mathbf{0}$. For this reason, a more general choice for $X(\mathbf{r})$ which fulfills the conditions in Eqs. (2) is

$$X(\mathbf{r}) = \rho_0(\mathbf{r} - u(\mathbf{R})\mathbf{n}_0(\mathbf{R}) + \mathsf{L}(\mathbf{r} - \mathbf{s}(\mathbf{R}))\mathbf{s}(\mathbf{R})), \tag{6}$$

where $\mathbf{R}\equiv\mathbf{R}(\mathbf{r})$ [see Eq. (3)]. As mentioned above, this study elucidates the general influence of $\mathsf{L}(\mathbf{r})$ on the height correlations. We will not derive its particular form. From the physical meaning discussed above in terms of tangential density fluctuations, it becomes clear that the integral kernel $\mathbf{L}(\mathbf{r};\mathbf{R},\mathbf{R}')$ should reflect the presence of an interfacial symmetry of the equilibrium interface $\mathbf{s}_0$. We will demonstrate this principle below in the case of planar interfaces.

An expansion of $X(\mathbf{r})$ given by Eq. (6) with respect to $u(\mathbf{R})$ up to linear order gives

$$X(\mathbf{r}) - \rho_0(\mathbf{r}) \approx -\nabla\rho_0(\mathbf{r})[\mathbf{1}_3 - \mathsf{L}(d\mathbf{n}_0(\mathbf{R}))]u(\mathbf{R})\mathbf{n}_0(\mathbf{R}), \tag{7}$$

where $\mathbf{1}_3$ means the 3×3 unit matrix, $d\equiv d(\mathbf{r})$ is the distance of $\mathbf{r}$ to the surface $\mathbf{s}_0$, and $\mathbf{R}\equiv\mathbf{R}(\mathbf{r})$ [see Eq. (3) and Fig. 1]. $\mathsf{L}(d\mathbf{n}_0(\mathbf{R}))u(\mathbf{R})\mathbf{n}_0(\mathbf{R})$ can be decomposed into normal and tangential contributions with respect to $\mathbf{s}_0(\mathbf{R})$ at each $\mathbf{R}$ which allows for a general derivation of the height correlations. Here, we demonstrate the main idea for *planar* interfaces. More general cases like the spherical or the cylindrical interface differ by their coordinate system which makes primarily the notation more complex. In addition, closed interfaces (e.g., droplets) are subject to an additional volume constraint for stability reasons; these exclude certain types of fluctuations. The crucial point in all cases is the symmetry of the equilibrium interface which is reflected by the correlation function $G(\mathbf{r},\mathbf{r}')$ and the integral kernel $\mathbf{L}(\mathbf{r};\mathbf{R},\mathbf{R}')$.

In the following discussion of height correlations of infinite planar interfaces [$\mathbf{R}\equiv(R_1,R_2)\in M=\mathbb{R}^2$], the equilibrium surface $\mathbf{s}_0(\mathbf{R})=R_1\mathbf{e}_x+R_2\mathbf{e}_y+z_0\mathbf{e}_z$, with a constant z_0, lies parallel to the (x,y) plane so that $\mathbf{n}_0(\mathbf{R})=\mathbf{e}_z$. The equilibrium density $\rho_0(\mathbf{r})\equiv\rho_0(z)$ depends only on the normal distance $z=z_0+d(\mathbf{r})$ to the surface $\mathbf{s}_0$ [see Eq. (3) and Fig. 1]. Consequently, the density correlation function $G(\mathbf{r},\mathbf{r}')\equiv G(z,z',\mathbf{R}-\mathbf{R}')$ is homogeneous with respect to the lateral coordinates. Equally, the integral kernel possesses the same homogeneity, i.e., $\mathbf{L}(d\mathbf{n}_0;\mathbf{R},\mathbf{R}')\equiv\mathbf{L}(z-c;\mathbf{R}-\mathbf{R}')$. Since $\rho_0(z)$ depends only on z, it is $\partial_x\rho_0=\partial_y\rho_0=0$ and Eq. (7) becomes

$$X(z,\mathbf{R}) - \rho_0(z) \approx -\partial_z\rho_0(z)[1 - \mathsf{L}(z - z_0)]u(\mathbf{R}), \tag{8}$$

where $\mathsf{L}(z-z_0)u(\mathbf{R})=\int_{\mathbb{R}^2}L(z-z_0;\mathbf{R}-\mathbf{R}')u(\mathbf{R}')d\mathbf{R}'$ with $\mathsf{L}(0)=0$ is a linear operator which depends on $z-z_0$ and which acts on the random variable $u(\mathbf{R})$ [see also Eq. (5)]. The correlation function for $X(z,\mathbf{R})-\rho_0(z)$ is given by $G(z,z',\mathbf{R}-\mathbf{R}')$. By taking the Fourier transformation with respect to the lateral coordinates, we obtain from Eq. (8)

$$\begin{aligned}2\pi\delta(\mathbf{q}+\mathbf{q}')\hat{G}(z,z',\mathbf{q}) = \partial_z\rho_0(z)\partial_z\rho_0(z')[1-\hat{L}(z-z_0;\mathbf{q})]\\ \times[1-\hat{L}(z'-z_0;\mathbf{q}')]\overline{\hat{u}(\mathbf{q})\hat{u}(\mathbf{q}')}.\end{aligned} \tag{9}$$

The interpretation of L as a projector of tangential density fluctuations on normal density fluctuations implies that $\hat{L}$ includes the ratio of Fourier components of the transverse autocorrelations and the height autocorrelation $\overline{\hat{u}(\mathbf{q})\hat{u}(\mathbf{q}')}$. Therefore and from the discussion above it becomes clear that, in particular for larger q values, $\hat{G}$ is reasonably governed by the transverse correlations and not only by the height correlations as in the case $\mathsf{L}\equiv 0$.

$\hat{G}(z,z',\mathbf{q})$ is a positive definite function and symmetric in z and z'. This allows for an expansion of Eq. (9) in terms of eigenfunctions of $\hat{G}(z,z',\mathbf{q})$, which leads to a generalized version of Wertheim's eigenfunction analysis of the correlations in a planar liquid-gas interface for small values of $|\mathbf{q}|$ [24]. The need of such a generalized approach has been already concluded from numerical investigations of the eigenfunction ansatz [25]. Here, we first ask in which way the term $\hat{L}(z-z_0;\mathbf{q})$ affects the *general form* of $\overline{\hat{u}(\mathbf{q})\hat{u}(\mathbf{q}')}$ which will be answered below *without* using an expansion in eigenfunctions. For low q values, the result becomes equal to the expression from Wertheim's approach. It is important to bear in mind that $\hat{G}(z,z',\mathbf{q})$ results from a thermodynamic average procedure while the height correlation function $\overline{\hat{u}(\mathbf{q})\hat{u}(\mathbf{q}')}$ stems from an unknown probability measure. Therefore Eq. (9) is in particular a manifestation of the required thermodynamic consistency condition mentioned above.

The *inverse* density correlation function $G^{-1}(\mathbf{r},\mathbf{r}')$ is defined by $\int d\mathbf{r}' G(\mathbf{r},\mathbf{r}')G^{-1}(\mathbf{r}',\mathbf{r}'')=\delta(\mathbf{r}-\mathbf{r}'')$. This implies from Eq. (9) for the height correlation function

$$\overline{\hat{u}(\mathbf{q})\hat{u}(\mathbf{q}')} = 2\pi\delta(\mathbf{q}+\mathbf{q}')C(\mathbf{q}) \tag{10a}$$

$$C(\mathbf{q}) := \left[\int\int_{-\infty}^{\infty} dz dz' \partial_z\rho_0(z)\partial_{z'}\rho_0(z')[1-\hat{L}(z-z_0;-\mathbf{q})] \times[1-\hat{L}(z'-z_0;\mathbf{q})]\hat{G}^{-1}(z,z',\mathbf{q})\right]^{-1}. \tag{10b}$$

G^{-1} is typically written as $G^{-1}(\mathbf{r},\mathbf{r}')=\delta(\mathbf{r}-\mathbf{r}')/\rho_0(\mathbf{r})-c(\mathbf{r},\mathbf{r}')$, where $c(\mathbf{r},\mathbf{r}')$ is called the direct correlation function [26]. In Fourier space, we have for the planar interfaces $\hat{G}^{-1}(z,z',\mathbf{q})=\delta(z-z')/\rho_0(z)-\hat{c}(z,z',\mathbf{q})$. Furthermore, the equilibrium density $\rho_0(z)$ fulfills the generalized barometric law, i.e., the equation $\ln\lambda_{\text{th}}^3\rho_0(z)-c^{(1)}(z)+\beta V^{\text{ext}}(z)=\beta\mu$, where λ_{th} is the thermal de-Broglie wavelength, $c^{(1)}(z)$ means the effective one-particle potential, $V^{\text{ext}}(z)$ is an external potential, and μ means the chemical potential [2]. By taking the derivative of the equilibrium condition for $\rho_0(z)$ and bearing in mind the relation $\nabla c^{(1)}(\mathbf{r})=\int d\mathbf{r}' c(\mathbf{r},\mathbf{r}')\nabla'\rho_0(\mathbf{r}')$ (see Ref. [2]) we can rewrite the ideal gas contribution in G^{-1}. This gives

$$\hat{G}^{-1}(z,z',\mathbf{q}) = -\beta\frac{\delta(z-z')\partial_z V^{\text{ext}}(z)}{\partial_z\rho_0(z)} - \hat{c}(z,z',\mathbf{q}) + \frac{\delta(z-z')}{\partial_z\rho_0(z)}\int dz''\hat{c}(z,z'',\mathbf{0})\partial_z\rho_0(z''). \tag{11}$$

By combining Eqs. (10) and (11) we obtain

$$\beta\overline{\hat{u}(\mathbf{q})\hat{u}(\mathbf{q}')} = \frac{2\pi\delta(\mathbf{q}+\mathbf{q}')}{v^{\text{ext}}(\mathbf{q})+\eta(\mathbf{q})}, \tag{12a}$$

where

$$v^{\text{ext}}(\mathbf{q}) := -\int_{-\infty}^{\infty} dz\partial_z V^{\text{ext}}(z)\partial_z\rho_0(z)|1-\hat{L}(z-z_0;\mathbf{q})|^2 \tag{12b}$$

and

$$\eta(\mathbf{q}) := \frac{1}{\beta}\int\int_{-\infty}^{\infty} dz dz' \partial_z\rho_0(z)\partial_{z'}\rho_0(z') \times\left\{[\hat{c}(z,z',\mathbf{0})-\hat{c}(z,z',\mathbf{q})]|1-\hat{L}(z-z_0;\mathbf{q})|^2 + \frac{1}{2}\hat{c}(z,z',\mathbf{q})|\hat{L}(z-z_0;\mathbf{q})-\hat{L}(z'-z_0;\mathbf{q})|^2\right\}. \tag{12c}$$

By construction, the height-height correlations given by Eqs. (12a)–(12c) are thermodynamically consistent with the density covariance function $G(\mathbf{r},\mathbf{r}')$. From the derivation above, the $\mathbf{q}$ dependence of $\eta(\mathbf{q})$ comes in via the direct correlation function $c(\mathbf{r},\mathbf{r}')$ and the kernel $\hat{L}(z;\mathbf{q})$ of the linear operator $\mathsf{L}(z)$. While $c(\mathbf{r},\mathbf{r}')$ represents the particle interactions, $\mathsf{L}(z)$ takes into account the change of the equilibrium density $\rho_0(z)$ due to a local volume change at point z, i.e., the local compressibility, or the ratio of tangential density correlations and normal density correlations, respectively.

The derivation of the explicit expression for $\overline{\hat{u}(\mathbf{q})\hat{u}(\mathbf{q}')}$, i.e., from Eqs. (9)–(11) and (12a)–(12c), is based on the definition for $\hat{G}^{-1}(z,z',\mathbf{q})$ and the generalized barometric law. Therefore *any choice* for $\hat{L}$ leads to a height correlation function [Eqs. (12a)–(12c)] which is consistent with the density covariance $G(\mathbf{r},\mathbf{r}')$. Putting Eq. (12a) back into Eq. (9) it follows that

$$\beta\hat{G}(z,z',\mathbf{q}) = \partial\rho_0(z)\partial\rho_0(z') \times\frac{[1-\hat{L}(z-z_0;\mathbf{q})][1-\hat{L}(z'-z_0;-\mathbf{q})]}{v^{\text{ext}}(\mathbf{q})+\eta(\mathbf{q})}, \tag{13}$$

where the functional dependence of $v^{\text{ext}}(\mathbf{q})$ and $\eta(\mathbf{q})$ on $\hat{L}(z-z_0;\mathbf{q})$ is given by Eqs. (12b) and (12c), respectively. Consequently, for a given $\hat{G}(z,z',\mathbf{q})$, Eq. (13) is a definition for $\hat{L}(z-z_0;\mathbf{q})$ [possibly limited by the linearization made in Eq. (8)]. For instance, using the total correlation function $\hat{h}(z,z',\mathbf{R}-\mathbf{R}')$, it is $\hat{G}(z,z',\mathbf{q})=\delta(z-z')\rho_0(z)+\rho_0(z)\rho_0(z')\hat{h}(z,z',\mathbf{q})$. From the Ornstein-Zernike relation for inhomogeneous systems combined with some closure relation, an approximate scheme for $\hat{h}(z,z',\mathbf{q})$ can be applied in order to get a functional equation for $\hat{L}(z-z_0;\mathbf{q})$. In reverse, a given $\hat{L}$ implies a particular form of $\hat{G}(z,z',\mathbf{q})$, where the physical meaning of $\hat{L}$ [as discussed above in Eq. (5)] might serve as a guidance. The corresponding height correlation function is in any case consistently given by Eqs. (12a)–(12c). That means in particular for larger q values that a sole investigation of $\overline{\hat{u}(\mathbf{q})\hat{u}(\mathbf{q}')}$, i.e., a sole examination of Eqs. (12a)–(12c), is not sufficient in order to check the reliability of a given $\hat{L}(z-z_0;\mathbf{q})$ [27].

The only approximations in the realization of Eqs. (2) are those in Eq. (6) or Eq. (7), respectively. Surprisingly, the concept of an effective interface Hamiltonian or an expansion of free energies, respectively, is not needed in order to derive the general structure of height correlations which are consistent with the underlying density correlations. Nevertheless, Eqs. (12a)–(12c) and in particular Eq. (12c) can be compared to the former expression for the height correlations based on such principles. To do so, it is convenient to consider $\gamma(q):=\eta(|\mathbf{q}|)/q^2$ which is referred to in literature as a wavelength dependent surface energy density for isotropic interfaces.

The macroscopic surface tension $\gamma(0)$ has been derived in Ref. [28] and follows from Eq. (12c), if $\hat{L}(z;\mathbf{q})\to 0$ for $q\to 0$. For steplike profiles $\partial_z\rho_0(z)\sim\delta(z-z_0)$ the $\hat{L}$ terms drop out due to $\hat{L}(0,\mathbf{q})=0$. In that case, Eq. (12c) reduces to the result derived in Ref. [4] if the same expression for the direct correlation function provided by the density functional in [4] is used [see Eq. (4.6) in Ref. [4]]. Both cases reflect some properties of $\hat{L}$ which we concluded from the general discussion above, i.e., the long-wavelength limit and the $\xi=0$ case.

Equations (12a)–(12c) include also the energy density for surface excitations derived by Mecke and Dietrich in Gaussian approximation [6]. To see that, we first note that the particular density functional used in [6] implies an explicit expression $\hat{G}_{\mathsf{MD}}^{-1}(z,z',q)$ for the inverse density covariance function and thus for the direct correlation function $\hat{c}_{\mathsf{MD}}(z,z',q)=\frac{\delta(z-z')}{\rho_0(z)}-\hat{G}_{\mathsf{MD}}^{-1}(z,z',q)$ [29]. In Ref. [6], the external potential is the homogeneous gravity potential $V_{\mathsf{MD}}^{\mathsf{ext}}(z)=mgz$ and the equilibrium interface is located at $z_0=0$ [see Eq. (2.4) in [6]]. By repeating the derivation of the Eqs. (12a)–(12c) from Eq. (10), one applies Eq. (11) with $V^{\mathsf{ext}}(z)\equiv V_{\mathsf{MD}}^{\mathsf{ext}}(z)$ and $\hat{c}(z,z',q)\equiv\hat{c}_{\mathsf{MD}}(z,z',q)$ except for the term $\partial_z\rho_0(z)\partial_{z'}\rho_0(z')\hat{L}^*(z;q)\hat{L}(z';q)\hat{G}^{-1}(z,z',q)$ which appears in Eq. (10). Therein, one uses $\hat{G}_{\mathsf{MD}}^{-1}(z,z',q)$ instead of Eq. (11) [which leads to the $\kappa-\tilde{\kappa}_0^{(HH)}(q)$ contribution in Eq. (3.11) in [6]]. By setting $\partial_z\rho_0(z)\hat{L}(z;\mathbf{q})\equiv-q^2\rho_H(z)$ [where ρ_H is given by Eqs. (3.27) and (3.31) in Ref. [6] without further derivation] the resulting expression for $\overline{\hat{u}(\mathbf{q})\hat{u}(\mathbf{q}')}$ and $\eta(q)/q^2$ are equal to Eqs. (4.1), (4.2), and (3.11), respectively, in Ref. [6] [see also Eqs. (2.28) and (3.10) in [6]]. From the explicit expression for $\rho_H(z)$ and $\rho_0(z)$ assumed in [6] we obtain $\hat{L}_{\mathsf{MD}}(z,\mathbf{q})=C_H/\pi\xi z\sinh(\frac{z}{2\xi})q^2$ [30]. Equation (13) implies that $\hat{L}_{\mathsf{MD}}(z,q)$ combined with $\hat{c}_{\mathsf{MD}}(z,z',q)$ provide rather a particular model for the density covariance function $\hat{G}(z,z',q)$ than a model for the height correlations only. Therefore in order to test the reliability of the resulting expressions in computer simulations it might be easier to check Eq. (13) instead of Eqs. (12a)–(12c) via arguable numerical procedures to define the position of the fluctuating interface.

The form of $\hat{L}_{\mathsf{MD}}(z,\mathbf{q})$ implies that the operator $\mathsf{L}_{\mathsf{MD}}(z)$ is not bounded for $z\neq0$ since $\mathsf{L}_{\mathsf{MD}}(z)\sim\nabla^2$ so that the kernel $L_{\mathsf{MD}}(z,\mathbf{R}-\mathbf{R}')$ involves the δ-distribution and its second derivative in the x and y directions. From Eq. (6) it follows that $X(\mathbf{r})\in[\rho_0^+,\rho_0^-]$, where $\rho_0^{\pm}:=\rho_0(z\to\pm\infty)$ denotes the bulk densities. Therefore a realization $u(\mathbf{R})$ of the interface with $u(\mathbf{R}_p)=|\nabla u(\mathbf{R}_p)|=0$ and $|\nabla^2u(\mathbf{R}_p)|=\infty$ for a particular point $\mathbf{R}_p$ induces $X(z\neq0,\mathbf{R}_p)=\rho_0^{\pm}$ [31]. On the other hand, the linearization of Eq. (6) [Eq. (8) for planar interfaces] with $\mathsf{L}\equiv\mathsf{L}_{\mathsf{MD}}$ allows for $X(\mathbf{r})\in[-\infty,\infty]$ even if $u(\mathbf{R})$ is bounded, which seems not reasonable from the physical point of view. Therefore as long as one would like to linearize Eq. (6), one has to limit the applicability of $\mathsf{L}_{\mathsf{MD}}(z)$ to a particular set of interface configurations whose second derivatives are also bounded, or equivalently, one has to limit the q range of the kernel $\hat{L}_{\mathsf{MD}}(z,\mathbf{q})$ [32].

The limitation of the set of interface configurations $\{u(\mathbf{R})\}$ restricts the set of modeled density configurations. If $\{X(\mathbf{r})\}$ denotes the set of all density configurations, an optimal sampling of $\{X(\mathbf{r})\}$ would capture the set of all *relevant* density configurations $\{X(\mathbf{r})\}_{\mathsf{rel}}\subseteq\{X(\mathbf{r})\}$ which, in our case, contribute to the density covariance function $G(\mathbf{r},\mathbf{r}')$. The representation of $X(\mathbf{r})$ by the interfacial field $u(\mathbf{R})$ leads to a set of density configurations $\{X(\mathbf{r})\}_{\mathsf{if}}$ which gives in general not the optimal sampling of $\{X(\mathbf{r})\}$ such that $\{X(\mathbf{r})\}_{\mathsf{if}}\subset\{X(\mathbf{r})\}_{\mathsf{rel}}$. Therefore one should allow for the *largest* set of interface configurations $\{u(\mathbf{R})\}$ (whose Fourier transform is continuous, for instance) in order to exhaust $\{X(\mathbf{r})\}_{\mathsf{rel}}$ in an optimal way, which corresponds to an approximation of $G(\mathbf{r},\mathbf{r}')$ by the height correlation function via Eq. (13) in an optimal way. As an example, one may allow for all interface configurations $u(\mathbf{R})$ with $\|u\|_1:=\int_{\mathbb{R}^2}|u(\mathbf{R})|d\mathbf{R}<\infty$ and in order to use Eq. (8), one may require $|L(z,\mathbf{R})|<\infty$ for all $\mathbf{R}\in\mathbb{R}^2$ so that $\mathsf{L}(z)$ becomes a bounded operator for every $|z|<\infty$ [33].

The operator $\mathsf{L}_{\mathsf{MD}}(z\neq0)\sim\nabla^2$ is associated to the *local* curvature of the interface $u(\mathbf{R})$ since it results from a truncated curvature expansion of the corresponding density configuration [see Eq. (2.17) in [6]]. Consequently, the coefficient of the resulting q^2 increase in $\gamma_{\mathsf{MD}}(q)$ has been interpreted as the bending rigidity of the liquid interface. The above considerations about the boundedness of $\mathsf{L}(z)$ then suggest that the influence of the local curvature of the interface and the effect of bending rigidity can be probably not continued to arbitrary small scales although the random density $X(\mathbf{r})$ as well as the random interface $u(\mathbf{R})$ *are* defined on all length scales. Nevertheless, since $\mathsf{L}(z)u(\mathbf{R})$ is written as a convolution integral [see Eq. (8)] and $\mathsf{L}(z)$ is bounded, we thus may imagine that $\mathsf{L}(z)$ picks up the *nonlocal* effects of the interface $u(\mathbf{R})$ on the density configuration $X(\mathbf{r})$. In a similar manner, the influence of nonlocality has been mentioned also in the framework of short-ranged wetting [34].

In our treatment of interface fluctuations, the distorted interface $\mathbf{s}(\mathbf{R})$ is considered to be generated by local *random* displacements $u(\mathbf{R})$ along the normal vector $\mathbf{n}_0(\mathbf{R})$ of the equilibrium interface $\mathbf{s}_0(\mathbf{R})$ [see Eq. (1) and Fig. 1]. By linking the random displacements to a change in density via Eq. (6), the density correlations $G(\mathbf{r},\mathbf{r}')$ govern the interface correlations $\overline{u(\mathbf{R})u(\mathbf{R}')}$. The situation is in a sense *reverse* to those in stochastic differential equations, where the mean value and the correlations of added noise terms [which correspond to $u(\mathbf{R})$ here] are specified in order to study its influence on a variable of interest [which is $\rho(\mathbf{r})$ in our case]. In general, the noise contribution within a single realization of the process cannot be determined unambiguously. Similarly, the difficulty to obtain $\overline{u(\mathbf{R})u(\mathbf{R}')}$ from numerical simulations arises from the problem to identify the random interface, i.e., the realization of the related (lower dimensional) stochastic process, in a particle configuration [22,23]. Consequently, any recipe that determines the random interface in computer simulations works accurately with a certain probability but cannot be exact. As pointed out in Ref. [19], the precision of such procedures is enhanced the more microscopic information of the system is included, i.e., the more n-particle correlations are taken into account to define the random interface numerically.

At this point we come back to the criticism of Tarazona, Checa, and Chacón [19] concerning the Mecke-Dietrich approach. In order to obtain the same expression for the height correlation function as published in [6] we have neither used an effective interface Hamiltonian nor a restricted variational principle as assumed in [6]. Formally, the random density

$X(\mathbf{r})$ corresponds to $\rho_f(\mathbf{r})$ in Ref. [6] [see Eq. (2.6) in [6]] but the crucial difference consists in the assumption that $\rho_f(\mathbf{r})$ in [6] minimizes the given density functional with an additional isodensity condition [see Eq. (2.5) in [6]]. While the isodensity condition for ρ_f in [6] is similar to the isodensity condition [Eq. (2a)] for $X(\mathbf{r})$, no additional equation resulting from a minimization procedure is required for $X(\mathbf{r})$, i.e., the realization of Eqs. (2) are those in Eq. (6) or Eq. (7), respectively. In order to extract the height correlation function or $\gamma_{\mathrm{MD}}(q)$, respectively, from computer simulations, the authors of Ref. [19] have taken this minimization condition for ρ_f in [6] seriously into account. Their results differ considerably from the predicted $\gamma_{\mathrm{MD}}(q)$ due to general features of density functionals as explained in Ref. [19]. Since ρ_f fulfills a minimization condition for a density functional, it includes capillary waves on small wavelengths which cannot be separated as height fluctuations (see Fig. 2 in [19]). While the numerical analysis shows that the minimization condition leads to a different $\gamma_{\mathrm{MD}}(q)$ than predicted, our approach shows that $\gamma_{\mathrm{MD}}(q)$ can be derived indeed *without* an additional (minimization) condition. In other words, the results in [19] do not necessarily imply that $\gamma_{\mathrm{MD}}(q)$ is structurally incorrect because no density functional and related minimization procedures are needed in order to derive $\gamma_{\mathrm{MD}}(q)$. The only dubious quantity that remains is $\hat{L}(z-z_0,q)$ or $\hat{L}_{\mathrm{MD}}(z,q)$, respectively, and from the derivation above it becomes clear that a model for $\hat{L}(z-z_0,q)$ is a different task that cannot be solved within a capillary wave theory. In particular, our derivation implies that $\hat{L}(z-z_0,q)$ or $\hat{L}_{\mathrm{MD}}(z,q)$ should be discussed rather in terms of Eq. (13) than only in terms of Eqs. (12a)–(12c). This suggestion if fully consistent with the conclusion in [19] that the capillary wave problem cannot be solved (numerically) on short wavelengths by taking into account only the one particle distribution.

Above we have discussed in detail the q-dependence of $\hat{L}(z,q)$ or $\hat{L}_{\mathrm{MD}}(z,q)$, respectively. Finally we briefly discuss a recently published study on planar colloid-polymer interfaces by Blokhuis, Kuipers, and Vink (BKV) [35] as an application of Eq. (13) (with $z_0=0$) in order to show that also the z dependence of $\hat{L}(z,q)$ plays a crucial role. In Ref. [35] a particular model for the so-called *surface density-density correlation function* $S(\mathbf{q}):=(\Delta\rho)^{-2}\int\int_{-\infty}^{+\infty}G(z,z',\mathbf{q})dzdz'$ [see Eq. (1) in Ref. [35] where $\Delta\rho:=\rho^{-}-\rho^{+}$ is the difference of the coexisting bulk densities] is proposed in order to explain the numerical data. $S_{\mathrm{BKV}}(\mathbf{q})$ in [35] contains an interfacial contribution $S_{hh}(\mathbf{q})$ and a bulk contribution $S_b(\mathbf{q})$ [see Eq. (11) in [35]]. Since $S_b(\mathbf{q})$ is modeled independently from $S_{hh}(\mathbf{q})$ [the bulk density fluctuations are considered to be *uncorrelated* from the interface fluctuations, see Eq. (10) in [35]] and only $S_{hh}(\mathbf{q})$ is used to adjust the data fit [Eqs. (14) and (15) in [35]], we focus here only on $S_{hh}(\mathbf{q})$. In order to compare $S(\mathbf{q})$ resulting from Eq. (13) to $S_{hh}(\mathbf{q})$, we adopt $v^{\mathrm{ext}}\equiv 0$ and use also the Helfrich form $\eta(\mathbf{q})/q^2=\sigma+q^2\kappa$ [see Eqs. (8) and (9) in [35]], where σ means the macroscopic surface tension and $\kappa\geqslant 0$ means the (phenomenological) bending rigidity. From Eq. (4) in [35] we identify $\partial_z\rho_0(z)\hat{L}_{\mathrm{BKV}}(z,\mathbf{q})=\frac{1}{2}\rho_1(z)q^2$, where $\rho_1(z)$ is given by Eq. (26) in [35]. Interestingly, the kernels $\hat{L}_{\mathrm{MD}}(z,\mathbf{q})$ and $\hat{L}_{\mathrm{BVK}}(z,\mathbf{q})$ show the same q dependence, but the weight functions $\rho_H(z)$ and $\rho_1(z)$, respectively, are different. In particular, it is $\rho_1(z=0)\neq 0$ so that the isodensity condition Eq. (2a) is not fulfilled. By evaluating the integral $\frac{1}{\Delta\rho}\int\partial_z\rho_0(z)[1-\hat{L}(z,\mathbf{q})]dz$, we then obtain from Eq. (13)

$$S(\mathbf{q})=\frac{(1+Cq^2)^2}{\beta\sigma q^2\left(1+\dfrac{\kappa}{\sigma}q^2\right)},\qquad(14)$$

with $C=C_H\xi^2$ (MD) or $C=0$ (BKV), respectively (see [30] and Eq. (6) in [35]). The best fit to the data in [35] *for the entire q range* results from Eq. (15) in [35] which implies $S_{hh}(\mathbf{q})=(\beta\sigma q^2)^{-1}(1+\frac{\kappa}{\sigma}q^2)$ with $\kappa=-\kappa_{\mathrm{BKV}}>0$ because κ_{BKV} is found to be negative (Table I in [35]). This form for $S_{hh}(\mathbf{q})$ is obtained from Eq. (14) for $\kappa=C\sigma$ if $C>0$ which is indeed the case for the $\hat{L}_{\mathrm{MD}}(z,\mathbf{q})$ but not for $\hat{L}_{\mathrm{BVK}}(z,\mathbf{q})$. Since Eq. (13) expresses the consistency between the height correlation function and the density covariance function, this result indicates that the kernel $\hat{L}_{\mathrm{MD}}(z,\mathbf{q})$ seems to be even more consistent than $\hat{L}_{\mathrm{BVK}}(z,\mathbf{q})$ with both, the assumed Helfrich form for $\eta(q)/q^2$ *and* the numerical results for $S_{hh}(\mathbf{q})$ *for the hole q range* in [35]. In particular, the *negative* bending rigidity κ_{BKV} in [35] appears rather artificially from the truncated Taylor expansion of $S_{hh}(\mathbf{q})$ in Eq. (14) in [35], which is assumed to be applicable for all q. Another main result of [35] is that κ_{BKV} vanishes for $T\to T_c$. Using $\kappa=C_H\sigma\xi^2$ from the relation above, it then follows that C_H vanishes with $T\to T_c$. This is in *qualitative* agreement with $C_H(T)\sim\xi^{-s}(T)$ for $s>0$ which has been concluded from general considerations about the expected quantitative influence of $\hat{L}_{\mathrm{MD}}(z,\mathbf{q})$ at higher temperatures [see comments on Eq. (55) in [7] and discussion on Eq. (4.72) in [36]], but without determining the exponent s rigorously [37]. Thus, although our approach does not provide a particular theory for $\hat{L}(z,\mathbf{q})$, it might serve as a tool in order to verify the consistency of various models with the underlying density covariance function.

X-ray experiments indicate $\hat{L}(z;\mathbf{q})\sim q^2$ for isotropic interfaces [9–14]; but a separation of the height correlations from the scattering data corresponds to Eq. (13). As discussed above, any choice for $\hat{L}$ leads to a height correlation function which is consistent with $\hat{G}$. This probably makes clear the uncertainty of such procedures at larger q values. Although a generalization of Eqs. (12a)–(12c) to multicomponent liquid mixtures as well as for spherical interfaces can be performed, the more challenging task is to identify the correlation ratios or the local compressibility, respectively, hidden in L.

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[30] From Eqs. (3.27) and (3.31) in Ref. [6] it is $\rho_H(z) = \frac{C_H \triangle\rho}{4\pi} z \sinh(\frac{z}{2\xi})/\cosh^2(\frac{z}{2\xi})$ and from Eqs. (3.26) and (3.30) in Ref. [6] it is $\partial_z \rho_0(z) = -\frac{\triangle\rho}{4\xi}/\cosh^2(\frac{z}{2\xi})$. In Ref. [6] it has been already argued that $C_H > 0$.

[31] An example of a function $f: \mathbb{R} \to \mathbb{R}$, which itself and its first derivative is bounded but whose second derivative is unbounded, is $f(x) = (1 - e^{-x^4})\sin(\frac{1}{x^2})$. In particular, it is $f(0) = f'(0) = 0$, $|f''(x \to 0)| \to \infty$ and the Fourier transform of $f(x)$ exists since $|f(x)|$ is integrable.

[32] It is possible to allow mathematically for unbounded random densities $X(\mathbf{r})$; but from the physical point of view follows that density configurations with $|X(\mathbf{r})| = \infty$ as well as the corresponding interface configurations occur with zero probability which results also in a limited q range of $\hat{L}_{\mathrm{MD}}(z,\mathbf{q})$.

[33] If there is a number $c(z) \in \mathbb{R}^+$ so that $|L(z,\mathbf{R})| \leqslant c(z)$ for all $\mathbf{R} \in \mathbb{R}^2$ and $|z| < \infty$ then $\|\mathsf{L}(z)u\|_1 \leqslant c(z)\|u\|_1 < \infty$.

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