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The Decay of Correlation of the Two-Particle Distribution Function in a Phase-Separating Layer and the Possibility of Spatial Phase Coexistence

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We study the decay of correlation of the two-particle distribution function in a plane phase separating layer (e.g., a liquid in coexistence with its vapor). We argue that the decay may be poorer in this special case than in the more general situation of interfaces of arbitrary shape. The clustering is shown to be weaker than $|x-y|^{-(d-2)}$, d the space dimension, in contrast to the more general situation. In particular, we show that this poor clustering is entirely restricted to the interface itself. This stronger result allows to prove as a by-product the nonexistence of a plane interface in two dimensions. Furthermore we make some remarks concerning the physical consequences like, e.g., the degree of particle number fluctuations and the behavior of the compressibility in the interface. The results do hold for two-particle potentials of short range.

KEY WORDS: Liquid–gas transition; interface region; long-range correlation of pair distribution function.

1. INTRODUCTION

An interesting topic of currently active research in fluid physics and more generally in statistical mechanics is the investigation of non-translation-invariant Gibbs states, which means in particular in the regime where several phases of a substance can spatially coexist, resp. where phase separation can occur in multicomponent systems. Of particular interest in this field is the behavior of the various local physical quantities in the interfacial region. While of interest in itself it is also of relevance in a broader context, namely, phase transitions of the first kind, which means usually, away from the critical point. Typical candidates are, e.g., the liquid–gas and liquid–solid transitions.

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While there is a host of rigorous results concerning interfaces in lattice models, especially in the Ising model, (cf., e.g., the many papers of Lebowitz and coworkers, Gallavotti, etc.; as a recent account see Refs. 20 and 21), there are relatively few exact results being accessible in the case of truly continuous systems (e.g., for the Widom–Rowlinson model[17]). As far as the liquid state in general is concerned the more recent state of the art is given in Refs. 18 and 19; an older reference is also Ref. 22.

One of the main questions which seems to be still at issue at the moment is to what extent a stable liquid–gas interfacial region can actually exist for the force of gravitation going to zero and the volume approaching infinity. There do exist several points of view which do either support the so-called capillary wave ansatz or the intrinsic local surface quantities concept (cf., e.g., Ref. 18, p. 79ff). As to this point we will make some sketchy remarks in Section 4 together with introducing a mechanism which might cure perhaps some of the nasty features occurring in this context.

As a last point we want to mention that our approach is not restricted to the liquid–gas phase transition or one-component systems. In the case of several components, as, e.g., in the W–R model, additional slight technical as well as computational modifications are, however, necessary, which shall be given elsewhere. The same applies to quantum liquids as, e.g., He_3–He_4 mixtures where Poisson brackets are replaced by commutators.

The basic building block for most of the physically relevant quantities of classical statistical mechanics of continuous systems is the pair-distribution function (see any good textbook of fluid physics, e.g., Refs. 1, 2, and 15). Hence it suffices usually to study the behavior of this quantity. One of the most significant effects on the behavior of physical properties as, e.g., compressibilities, specific heats, free energy, etc., is produced by the degree of long-range correlation of this quantity.

This influence is well known at the critical point, where it is displayed in the form of the critical opalescence. Furthermore, concerning classical crystals it was shown by Mermin that large fluctuations in the particle density, in other words, the well-known $|k|^{-2}$ singularity in the Fourier spectrum, prevents the system from crystallizing in two dimensions (Ref. 3).

If one wants to deal with the regime where phase separation can exist the usual methods which work well in the case of pure homogeneous phases do not apply. A crucial preassumption of the applied techniques is an at least rudimentary form of translation invariance. In two dimensions there is however a more recent paper of Fröhlich and Pfister (Ref. 4) which does not use homogeneity but relies on free-energy estimates. The method however is sensible only in $d = 2$.

A different approach to treat phase transitions in inhomogeneous systems (i.e., not invariant under translations, resp. rotations), was developed independently by Ph. A. Martin, Ch. Gruber, and coworkers, and the author in a series of papers. The two approaches have the advantage of working in any dimension and of giving sensible results for the distribution functions. While the above-mentioned authors started from the BBGKY hierarchy we, on the other side, exploited the so-called Kubo–Martin–Schwinger relation $\langle\{A,B\}\rangle = \beta\langle B\cdot\{A,H\}\rangle$ (cf. Refs. 5–9), where Ref. 9 yielded also results for the dynamical regime.

A common feature of the above-cited papers was that by not assuming a rudimentary form of translation invariance, (e.g., under a Bravais lattice), a clustering of the two-particle, resp. three-particle, correlation functions only $\gtrsim c|y_1 - y_2|^{-(d-1)}$ was necessary if translation invariance was to be broken, (a short-range interaction assumed). On the other hand, for homogeneous pure phases below T_c it was well known for quite some time and for many special systems that the clustering is usually $\gtrsim |y_1 - y_2|^{-(d-2)}$. A general coordinate space proof of this result was given recently in Ref. 10.

But one should emphasize that a system consisting of, e.g., finite liquid drops in coexistence with their vapor is quite distinct from, e.g., an ideal crystal phase, extending homogeneously to infinity. In the pure gas-phase, resp. liquid-phase, we expect the correlations to decay rather fast; the weak decay has to be attributed to the existence of the interfaces themselves. So there is no *a priori* reason to expect an analogous cluster behavior, in particular, the shape of the interfaces might play an important role. This is supported by the well-known difference of the vapor pressure as compared with the pressure inside the liquid drops, and which vanishes in case of a plane interface.

In the following we want to treat the special but important case of a plane interfacial layer, separating, e.g., a liquid from its vapor phase. We shall rigorously show in Chapter 3 that the clustering of the two-particle distribution function, more specifically $\rho_2^T(y_1, y_2) := \rho_2(y_1, y_2) - \rho_1(y_1)\cdot\rho_1(y_2)$, is extremely poor in the interface region itself, that is, the decay is weaker than $|y_1 - y_2|^{-(d-2)}$, d the space dimension, for $|y_1 - y_2| \to \infty$ in the interface. This confirms, extends, and quantifies the result of a recent interesting paper by M. S. Wertheim, who attacked this problem with the help of the direct correlation function method.[11] As a by-product we prove the nonexistence of plane interface regions in two dimensions. In Chapter 4 we shall discuss the physical consequences which are automatically implied by this poor decay because of the pair distribution functions occurrence in almost all physically relevant quantities.

As a final remark we want to mention a preprint of H. Narnhofer,[16] which we received after having finished this paper. There it is argued that there might occur difficulties in generating interfaces within the usual conceptual frame of statistical mechanics. On the other hand, the handling and interchanging of various limiting procedures in the limit $V \to \infty$ is crucial in this context. So, in our view, this point needs further clarification since exactly these long-range effects might invalidate the argumentation.

2. CONCEPTS AND TERMINOLOGY

The infinite phase space is the space of countable sequences $X := \{x_i\}$, $x_i = (q_i, p_i)$, $q_i, p_i \in R^d$. Observables are smooth localized functions on phase space, e.g., for an n-particle quantity: $A(X) = \sum_{\{i_1, \ldots, i_n\}} a(x_{i_1}, \ldots, x_{i_n})$, the summation extending over all ordered n-tuples, $a(\cdot)$ a smooth symmetric function on $(R^{2d})^n$. The Poisson bracket of A, B reads

$$\{A, B\}(X) := \sum_i (\partial A/\partial q_i \cdot \partial B/\partial p_i - \partial A/\partial p_i \cdot \partial B/\partial q_i) \tag{1}$$

Remark. More details can be found in the above-mentioned papers.

The expectation of A over the phase space is denoted by $\langle A \rangle$. A particularly important quantity is the particle density at site y:

$$n(y)(X) := \sum_i \delta(y - q_i) \tag{2}$$

and its expectation $\langle n(y) \rangle = \langle \sum_i \delta(y - q_i) \rangle$. The n-particle distribution function $\rho_n(y_1, \ldots, y_n)$ is defined by

$$\rho_n(y_1, \ldots, y_n) := \sum_{\{i_\nu\}} \langle \delta(y_1 - q_{i_1}) \cdots \delta(y_n - q_{i_n}) \rangle \tag{3}$$

in particular, $\langle n(y) \rangle = \rho_1(y)$. Furthermore, we define the truncated two-particle correlation function:

$$\rho_2^T(y_1, y_2) := \rho_2(y_1, y_2) - \rho_1(y_1)\rho_1(y_2) \tag{4}$$

with $\rho_2^T(y_1, y_2) \to 0$ for $|y_1 - y_2| \to \infty$ in a definite pure phase.

The various pure phases of the system with given macroscopic parameters are assumed to be generated in the thermodynamic limit via a procedure like the Bogoliubov quasi-average method, and by which measure, in particular, the phase boundaries are assumed to be given a definite position. It is however not clear whether this will work in this context, (cf., e.g., Ref. 20 and the remarks in the introduction), and for what space dimension of the system.

While in the more general situations mentioned in the introduction one has to rely on other methods, we are in the following able to apply the

Bogoliubov inequality of classical statistical mechanics, which reads

$$|\langle\{A,B\}\rangle|^2 \leqslant \beta\langle\{B,\{B,H\}\}\rangle\langle A\cdot A\rangle \tag{5}$$

with A,B real quantities.

3. THE DECAY OF CORRELATION IN THE INTERFACE

The following calculation is made for the interesting space dimension $d=3$, but is actually independent of it. The case $d=2$ will be treated in a corollary at the end of the paper.

The particles are assumed to interact via a two-body potential, $V(q) = V(-q)$, of short-range type. We will need it in the following technical form:

$$\int |\partial_i^2 V(q)|\,|q|^2\,dq < \infty$$

However, only the long-range behavior of the force is of relevance. That is, stronger singularities at $q=0$ are allowed on physical grounds and which are usually compensated for by the vanishing of other quantities at coinciding points [cf. expression (22)]. On the other hand a true hard core might cause certain difficulties, as was already mentioned in Ref. 3.

For notational simplicity we assume the interface region to be parallel to the x-y plane with $z=0$ to belong to this interfacial layer. Furthermore, we assume complete translation invariance with respect to the x-y directions. The invariance under a Bravais lattice can be treated in complete analogy. As observable A in (5) we take the particle density $n(y)$, B is generated by the density of the generator of translations in the z direction (for notational simplicity x, y, z as components of the vector y are in the following sometimes denoted y^1, y^2, y^3 to avoid confusion with the full position variables as, e.g., $y_1, y_2, \ldots$).

The physical idea behind our approach is the following: If there exists an interfacial region this implies a $\rho_1(z) \neq \text{const}$, resp. $\partial_z\rho_1(z) \neq 0$ for certain z values. This is exploited in (6). The detailed behavior of $\rho_1(z)$ may be complicated (perhaps even oscillating as is the case in certain model calculations in Ref. 15 Chapter 4), but this property is the only relevant one in the following. The next step in our strategy is to build in properly the translation invariance with respect to the x-y plane into the calculations. This is accomplished by replacing the local observable $n(y)$ by a certain mean with respect to the x-y coordinates [cf. (8)].

This method was applied recently in Ref. 10, resp. Ref. 12 in the context of systems invariant under a full Bravais lattice. It was also exploited in the realm of quantum field theory in connection with the Goldstone theorem, (cf. Refs. 13 and 14). That is, our personal contribution is the observation that a modification of these methods can be successfully applied to the physically important situation of phase boundaries and phase transitions of the first kind as, e.g., liquid–gas which was originally not accessible to these exact methods. The same does apply to the calculation of the double Poisson brakets $\langle\{P,\{P,H\}\}\rangle$ which we reproduce in this paper for reasons of self-containment and since the calculations are lengthy and a little bit tricky.

We have

$$-\partial_z\langle n(y)\rangle = \left\langle \left\{ n(y), \sum_i p_i^3 \right\}(X) \right\rangle = \left\langle \left\{ n(y) - \langle n(y)\rangle, \sum_i \cdots \right\} \right\rangle \tag{6}$$

We assume without loss of generality that $\partial_z\langle n(y)\rangle \neq 0$ for $y^3 = 0$. Furthermore, for various reasons (cf. also Refs. 5 and 10), we replace the full generator $\sum p_i^3$ by a localized version $P_R^3 := \sum p_i^3 \cdot f_R(q_i)$ with

$$\begin{aligned} f_R(y) &= f(|y|/R), \qquad && f \text{ being smooth with } f(s) = 1 \text{ for } s < 1, \\ &= 0 && \text{for} \quad s > 2. \end{aligned} \tag{7}$$

Evidently, the expression (6) is not changed by this replacement provided that $|y| < R$.

To exhibit the poor clustering in the y^1-y^2 plane we will use instead of $\delta n(y) = n(y) - \langle n(y)\rangle$ the following mean value on the right-hand side of (6):

$$A_R := 1/|V_R| \int \delta n(y) \cdot \chi_R(\bar{y})\, d\bar{y} \tag{8}$$

with χ_R the characteristic function of the set $V_R = \{\bar{y} \,|\, (y^1)^2 + (y^2)^2 < R^2\}$ and $\bar{y} := (y^1, y^2)$. Again, owing to the translation invariance in the x-y plane the left-hand side of (6) will not be altered by replacing $n(y)$ by the corresponding mean with respect to y^1, y^2.

The Bogoliubov inequality with these observables A_R, P_R^3 now reads

$$0 \neq |\partial_z\langle n(y)\rangle_{z=0}|^2 \leqslant \beta\langle\{P_R^3, \{P_R^3, H\}\}\rangle \cdot \langle A_R \cdot A_R\rangle \tag{9}$$

We will calculate the R dependence of the two expressions on the right-hand side separately. We have

$$\begin{aligned} \langle A_R \cdot A_R\rangle &= 1/|V_R|^2 \int\int d\bar{y}\, d\bar{y}'\, \chi_R(y)\chi_R(y') \\ &\quad \times (\langle n(y)n(y')\rangle - \langle n(y)\rangle\langle n(y')\rangle) \\ &= 1/|V_R|^2 \int \rho_2^T(\bar{x},0) \cdot |V_R \cap V_R^{\bar{x}}| \end{aligned} \tag{10}$$

We remind the reader that $y^3 = y'^3 = 0$, $\bar{x} = \bar{y} - \bar{y}'$; $|V_R \cap V_R^{\bar{x}}| = \int d\bar{y}\, \chi_R(y) \cdot \chi_R(y - x)$ is the volume of the intersection of V_R and V_R shifted by the vector $\bar{x}$.

With $|V_R \cap V_R^{\bar{x}}| \leqslant |V_R|$ and being $= 0$ for $|\bar{x}| > 2R$ we have the upper bound:

$$\langle A_R \cdot A_R \rangle \leqslant 1/|V_R| \int_{V_{2R}} d\bar{x}\, |\rho_2^T(\bar{x})| \tag{11}$$

Both on physical and mathematical grounds the truncated two-point function should cluster for $|\bar{x}| \to \infty$. Assuming for example that $|\rho_2^T(\bar{x})| < C \cdot (1 + |x|)^{-\alpha}$ with $\alpha > 0$, we arrive at the expression

$$\langle A_R A_R \rangle \leqslant C_1 R^{-\alpha} + C_2 R^{-2} \tag{12}$$

with C_1, C_2 appropriately chosen ($d = 3$), that is, for poor clustering (i.e., $\alpha < 2$), the dominating term behaves $\sim R^{-\alpha}$. We want to emphasize however that a preassumption about the degree of clustering is not really essential in the following. For $d = 2$, in particular, the weakest possible cluster behavior turns out to be already sufficient.

We now have to calculate the expression $\langle \{ P_R^3, P_R^3, H \} \} \rangle$. The inner bracket yields

$$\left\{ \sum_j p_j^3 \cdot f_R(q_j), H \right\} = \left\{ \sum_j p_j^3 \cdot f_R(q_j), \sum_i (p_i)^2/2m + 1/2 \sum_{k \neq 1} V(q_k - q_1) \right\}$$

$$= 1/m \sum_j \left(p_j^3\right)^2 \partial_{jj}^3 f_R(q_j) - \sum_{j \neq k} f_R(q_j) \partial_j^3 V(q_j - q_k) \tag{13}$$

Performing the other Poisson bracket yields two terms; the first reads

$$1/m \left(\sum_j \left(p_j^3\right)^3 \left(\partial_{jj}^3 f_R(q_j)\right)^2 - f_R(q_j) \left(\partial_j^3\right)^2 f_R(q_j) \left(p_j^3\right)^2 \right) \tag{14}$$

the second

$$\sum_{j \neq k} \left(\partial_j^3\right)^2 V(q_j - q_k) \left(f_R^2(q_j) - f_R(q_j) f_R(q_k) \right)$$

$$+ \sum_{j \neq k} \left(\partial_j^3\right) V(q_j - q_k) f_R(q_j) \partial_{jj}^3 f_R(q_j) \tag{15}$$

where $(\partial_i) V(q_j - q_i) = -(\partial_j) V(q_j - q_i)$ has been used.

Performing the expectation values yields

$$\left\langle \sum_j \left(p_j^3\right)^3 \left(\partial_{jj}^3 f_R(q_j)\right)^2 \right\rangle = 0 \tag{16}$$

since the momenta are distributed according to a Maxwellian which is not affected by exterior fields, fixing the phase boundary, and which do act only on the coordinates. Furthermore we have

$$0 = \left\langle \left\{ H, \sum_j p_j^3 f_R(q_j) \partial_j^3 f_R(q_j) \right\} \right\rangle \tag{17}$$

since we are in an equilibrium state. This yields

$$\left\langle \sum_{j \neq k} (\partial_j^3) V(q_j - q_k) f_R(q_j) \partial_j^3 f_R(q_j) \right\rangle - 1/m \left\langle \sum_j (p_j^3)^2 f_R(q_j) (\partial_j^3)^2 f_R(q_j) \right\rangle$$

$$= 1/m \left\langle \sum_j (p_j^3)^2 (\partial_j^3 f_R(q_j))^2 \right\rangle \tag{18}$$

The left-hand side occurs in $\langle \{ P_R^3, \{ P_R^3, H \} \rangle$, hence we are left with

$$\langle \{ P_R^3, \{ P_R^3, H \} \rangle = 1/m \left\langle \sum_j (p_j^3)^2 (\partial_j^3 f_R(q_j))^2 \right\rangle$$

$$+ \left\langle \sum_{j \neq k} (\partial_j^3)^2 V(q_j - q_k) \left(f_R^2(q_j) - f_R(q_j) f_R(q_k) \right) \right\rangle \tag{19}$$

With the help of the distribution functions this can be written as

$$\int \int (\partial_{y_1}^3)^2 V(y_1 - y_2) \left(f_R^2(y_1) - f_R(y_1) f_R(y_2) \right) \rho_2(y_1, y_2) \, dy_1 \, dy_2$$

$$+ 1/m \cdot \beta^{-1} \int dy \, (\partial_y^3 f_R(y))^2 \rho_1(y) \tag{20}$$

where β^{-1} comes from the integration over the momenta. Owing to the symmetry of ρ_2 with respect to y_1, y_2 and $(\partial_{y_1}^3)^2 V = (\partial_{y_2}^3)^2 V$ we can give the integrand in the first integral the more symmetric shape: $1/2 (\partial_{y_1}^3)^2 V(y_1 - y_2)(f_R(y_1) - f_R(y_2))^2$.

We will now take advantage of the special form of the function $f_R(y)$ given in (7). The second integral of (20) can be estimated as follows:

$$\int dy \, (\partial_y^3 f(y/R))^2 \rho_1(y) = R \cdot \int dy' \, (\partial^3 f(y'))^2 \rho_1(Ry') < C \cdot R \tag{21}$$

since ρ_1 is bounded. In the first integral we get

$$\left| \int dy \, (\partial_y^3)^2 V(y) \left(\int dy_2 \, (f_R(y + y_2) - f_R(y_2))^2 \rho_2(y + y_2, y_2) \right) \right|$$

$$< \int dy \, |(\partial_y^3)^2 V(y)| \left(\int dy_2 \, (f_R(y + y_2) - f_R(y_2))^2 \cdot \sup_{y_2} \rho_2(y + y_2, y_2) \right) \tag{22}$$

Furthermore

$$|f(y+y_2/R)-f(y_2/R)|=\left|\int_{y_2/R}^{y+y_2/R}\partial f\cdot d\mathbf{n}\right|\leqslant \sup|\partial f|\cdot|y|/R \quad (23)$$

The y_2 support of $(f_R(y+y_2)-f_R(y_2))^2$ for y fixed has a volume $\leqslant C'R^3$. Putting these remarks together we arrive at

$$\text{"rhs of (22)"} < C''R^3\int dy\left|\left(\partial_y^3\right)^2 V(y)\right|\cdot|y|^2/R^2 \leqslant \text{const}\cdot R \quad (24)$$

for short-range potentials as defined at the beginning of Chapter 3.

We have thus arrived at the main conclusion of this paper:

Theorem. Assuming a short-range potential, a clustering of the truncated two-particle correlation function $|\rho_2^T(y_1,y_2)|\leqslant C\cdot(1+|y_1-y_2|)^{-\alpha}$ in the interfacial layer, we have in three dimensions:

$$|\langle\{P_3,n(y)\}\rangle|^2\leqslant \lim_{R\to\infty}\text{const}\cdot R^{1-\alpha}$$

where P_3 is the generator of translations in the direction perpendicular to the interface. That is, if $\partial_z n(y)\neq 0$ somewhere then $\rho_2^T(y_1,y_2)$ has a decay in the interface which is $\gtrsim C\cdot|y_1-y_2|^{-1}$!

Corollary. For two space dimensions there is no liquid–gas interface provided that there is any clustering at all of ρ_2^T (which is of course physically desirable).

Sketch of Proof. Since the dimension of coordinate space played a role only in the estimate (24) (prefactor R^3 for $d=3$ which becomes R^2 for $d=2$), we can bound the left-hand side of (24) by a constant for $d=2$. Furthermore, for $d=2$, the weakest form of cluster behavior in a pure phase turns out to be sufficient in the expression $\langle A_R\cdot A_R\rangle$. With the translations parallel to the interface being unbroken, a general theorem says (cf. Ref. 23, p. 155), that suitable group means of observables with respect to this subgroup display "weak clustering." This entails in our case (remember that A_R, according to definition, is already an observable normalized with respect to its mean value!), that $\lim_{R\to\infty}\langle A_R\cdot A_R\rangle=0$. That is, for $d=2$ we have always $\langle\{P_3,n(y)\}\rangle=0$!

Remarks. (i) The sensible expression against, e.g., the formation of interfaces appears to be the quantity $\langle A_R A_R\rangle$! Only there appear the *truncated* two-particle distribution function and a domain of integration which takes the existence of the interface explicitly into account.

(ii) The expression $\langle\{P_R^3,\{P_R^3,H\}\}\rangle$ on the other side was completely general. Choosing a different domain of integration does not lead to better results. In particular, only the full distribution functions show up which do not decay!

4. PHYSICAL CONSEQUENCES

We have shown that there is a particularly poor clustering in the interface region. We will now make a couple of comments concerning the physical and observational consequences of this weak decay. Let us choose a subvolume V lying in the phase separating layer with a fixed but macroscopic extension in the z direction, the x-y extension being variable. Standard statistical mechanics tells us that the particle number fluctuations in V can be expressed as

$$\left\langle (N - \langle N \rangle)_V^2 \right\rangle = \int_V \int_V \rho_2^T(r, r')\, dr\, dr' \tag{25}$$

and the isothermal compressibility:

$$K_T = \rho^{-1} \cdot \beta \lim_{V \to \infty} \left\langle (N - \langle N \rangle)_V^2 \right\rangle / \langle N_V \rangle \tag{26}$$

where in (26) we assume V to approach ∞ in the x-y direction.

Usually the above double integral is transformed into $V \cdot \int_V \rho_2^T(r)\, dr$ which is only approximately correct for a sufficiently large V and a correlation extending only over several interatomic distances. Under this assumption the particle number fluctuations ΔN will behave $\sim V^{1/2}$ provided that ρ_2^T is integrable. In our case ρ_2^T is not of short range, even worse, the above integral may be divergent for $V \to \infty$ as at the critical point. There are exactly two alternatives. Either the above integral over ρ_2^T goes to infinity for $V \to \infty$ in the interface or ρ_2^T displays a sufficiently oscillatory behavior (which is known for the first interparticle distances) so that the Riemann integral may exist in the limit while ρ_2^T not being absolutely summable.

The first alternative would imply strong fluctuations in the interface which is not implausible since one can imagine that there is a permanent considerable amount of diffusion of, e.g., liquid drops into the vapor phase and vice versa. This would yield a ΔN_V larger than $\sim V^{1/2}$ which may cause phenomena in the surface similar to critical opalescence and a very large local compressibility. On the other hand, a sufficiently strong oscillation of ρ_2^T may provide a mutual compensation of the particle number fluctuations such that a normal $\Delta N \sim V^{1/2}$ would be restored. Up to now we have not found experimental evidence supporting one or the other possibility in the literature, but these experiments are obviously difficult to perform in the interfacial layer.

The derived results can now be interpreted in various ways. Either, as was already indicated in the introduction, there are no *stable* interfaces for $d = 3$ in a continuous system. Then the large fluctuations may just indicate this instability of the interface. Or there are large fluctuations *and* a *stable* interface. The third alternative would be an oscillatory decay of correlations so that the particle number fluctuations in macroscopic volumes remain normal.

While the first alternative is supported by the behavior of two-dimensional membranes and some lattice models there are at the moment no rigorous results for three-dimensional continuous models. In particular, we want to emphasize that the possible mechanism alluded to in the third alternative is typically absent in lattice models, where we have usually a nonoscillatory decay of correlations (and being displayed in the second Griffith inequality). Therefore we are not completely convinced that lattice models are reliable candidates in *every* respect.

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