Journal of Statistical Physics, Vol. 45, Nos. 5/6, 1986

Poor Decay of Correlations in Inhomogeneous Fluids and Solids and Their Relevance for the Physics of Phase Boundaries

M. Requardt[1] and H. J. Wagner[1]

Received March 10, 1986; revision received July 17, 1986

We study the poor decay of correlations for equilibrium states of inhomogeneous fluids and solids, in the regimes of both classical and quantum statistical mechanics. Our main observation is the usefulness of the statistical mechanical expression of the *stress tensor* and its long-range correlations with the particle density. From this we are able to infer a very slow decay of correlations for the various molecular distribution functions under discussion. The derived results are of relevance both for completely inhomogeneous systems such as quasicrystals or granular structures and for the slightly more regular cases of, e.g., phase separating layers in fluids and solids, ideal crystals, etc. As one of the byproducts we prove the nonexistence of plane *quantum* interfaces in two dimensions (thus extending earlier results of Requardt to the quantum regime). The results hold for arbitrary potentials of not too long range.

KEY WORDS: Inhomogeneous equilibrium states; phase boundaries; stress tensor; poor decay of correlation; static susceptibilities.

1. INTRODUCTION

While most of the bulk properties of classical (quantum) statistical systems are understood at least in principle—even their phase transitions and critical point behavior—and while in the last decade many exact results have been derived for classical lattice systems in the regime of spatial phase coexistence (cf., e.g., the many papers by J. L. Lebowitz and his group), there has been less progress in understanding the formation of non-translation-invariant Gibbs states of infinite continuous systems in the absence of exterior fields, especially interfaces between various substances or phases

[1] Institut für Theoretische Physik der Universität Göttingen, D-3400 Göttingen, West Germany.

0022-4715/86/1200-0815$05.00/0

and modifications of a single substance in the regime where they can spatially coexist. (As an aside we would like to add the remark that, contrary to a perhaps widespread belief, there are marked differences between continuous and lattice models, in particular concerning the phenomena we are referring to).

While, e.g., the statistical mechanical theory of surface tension dates back to the days of van der Waals, it was only quite recently understood that various of the tacitly made underlying assumptions are not correct or have to be modified. As far as the liquid–vapor interface is concerned, a recent account of the state of the art was given in the beautiful book of Rowlinson and Widom.[1] Other aspects have been discussed in, e.g., Refs. 2 and 3, the latter also treating systems like liquid metals and some quantum systems.

However, there seems to be much less work on quantum continuous systems, particularly papers discussing classical and quantum systems on more or less the same footing. Furthermore, whereas there are certain differences concerning the problem of, e.g., solid–liquid, solid–gas coexistence as compared to the liquid–vapor situation (cf., e.g., Ref. 4) most of the analysis can be done along the same lines and after some modifications also for quantum continuous systems.

One of the main differences as compared to phase transitions in the bulk is the role the exterior thermodynamic state fixing field plays in this context, which in the process of quasiaveraging is assumed to be switched off after the thermodynamic limit has been taken. There are arguments that for, e.g., space dimension $d \leqslant 3$ the interfacial layer starts to oscillate when the gravitational field goes to zero such that an interface may be defined only locally but not globally. This is, however, a subtle point, since the oscillations go only with the log of the surface area and the reasoning rests on several assumptions which are both crucial and difficult to verify, such as analyticity of the Fourier transform of the direct correlation function $c^{(2)}(r_1, r_2)$ in the transverse direction (cf., e.g., the remarks in Refs. 1, 5, and 6).

What can, however, rigorously be proved is that there are long-range correlations in the two-particle correlation function

$$\rho_T^{(2)}(r_1, r_2) := \rho^{(2)}(r_1, r_2) - \rho^{(1)}(r_1) \cdot \rho^{(1)}(r_2)$$

in the interface of the type $\lesssim |(r_1 - r_2)^{\perp}|^{-(d-2)}$ if a stable plain interface exists at all (cf. Ref. 7). This may have certain consequences for various physical quantities, such as the compressibility in the interfacial layer, and for the statistical mechanical expression of surface tension, some of which have been discussed in Refs. 7 or 8. But we would like to emphasize that one only gets information about the long distance behavior of $|\rho_T^{(2)}(r_1, r_2)|$

and *not* of $\rho_T^{(2)}(r_1, r_2)$ itself. Since $\rho_T^{(2)}$ *oscillates* around zero as a function of $(r_1 - r_2)$, integrals over terms containing $\rho_T^{(2)}$, as in expressions for the surface tension, might therefore possibly exist in a generalized Riemann sense while they diverge with $\rho_T^{(2)}$ substituted by $|\rho_T^{(2)}|$.

In any case some expressions of physical relevance may become nearly infinite in the interface, which may be interpreted either as meaning that there is no stable interface at all or that the interfacial layer displays certain quasicritical properties. On the other side, we have some difficulty in imagining that the interface between, e.g., a solid and a gas would actually wildly fluctuate in the absence of a gravitational field. Since at least our approach in Ref. 7 also covers experimental setups like these, we are not entirely convinced that there is only *one* universal interpretation of the phenomenon.

The problem of spatial phase coexistence can be approached on roughly two levels of complexity. One can assume either that a certain residual space symmetry survives the process of phase separation or that the inhomogeneous equilibrium state does not support some sort of restricted invariance group. A typical candidate of the first kind is a plane interface separating two phases like a liquid and its vapor or a solid and a liquid. In the latter case $\mathbb{R}^d$ is broken up into small fractions of the various phases or modifications of one or several substances. Well-known examples are superconductors of the second kind with their filament structures, and formation of Weiss domains in magnets or quasicrystals. It depends on the balance between bulk and surface free energy which state is actually favored.

According to this we divide the treatment into four parts. We start in Section 3 with the general inhomogeneous situation of classical statistical mechanics of continuous systems, i.e., we do not exploit any symmetry of a certain subgroup of space translations. We continue in Section 4 with the corresponding quantum case. In Section 5 we discuss the special but important situation of a planar interfacial layer both for classical and quantum continuous systems. What we are after in all these different setups is to display the linkage between the various degrees of breaking of translation invariance (resp. existence of density gradients) and long-distance correlations between certain relevant physical quantities and the resulting physical implications of this.

To this end, Section 2, apart from fixing notations and terminology, introduces the notion and properties of the statistical mechanical expression of *stress tensor*, in both the classical and quantum regimes. It is the stress tensor that turns out to be the relevant notion in this context. Since, espcially in the quantum case, a derivation from first principles of this quantity is given seldomly or only in some approximative way, we take the chance to give our own, self-contained derivation, which may be of use in itself.

As to Section 3, there already exist rigorous papers attacking the problem from various directions. In Ref. 9 the case of general spontaneous symmetry breaking in classical continuous systems was discussed, including space translations. In Ref. 10 the special case of breaking of translation and rotation invariance was treated for a wider class of potentials. While in Ref. 9 the classical Kubo–Martin–Schwinger property was used, Ref. 10 exploits the BBGKY hierarchy. These two concepts are related to each other, but possibly not completely equivalent (cf., Ref. 11). Our, as we hope, new contribution is to bring the stress tensor into play, thus making the physical implications and the reasoning much more transparent. Furthermore, the section serves as a warmup exercise for Section 4, where the corresponding situation is discussed for the quantum case.

It is characteristic that in the case of quantum continuous systems the expectation value of the kinetic part of the stress tensor is not trivial, in contrast to the classical equilibrium case. In particular, the off-diagonal terms of the closely related pressure tensor do not vanish, which reflects the nontrivial coupling of fluctuations in the various momentum components with each other and with the particle density. Furthermore, both the expressions we start from and the calculation in between differ from the ones in the classical case. The final results, however, are similar.

In Section 5 the special situation of a planar interfacial layer is treated for the quantum regime. It is typical that the results are stronger in this case due to the residual translation invariance in the directions parallel to the interface. Since the case of a one-component classical system was already treated in Ref. 7, we make only a short aside concerning a special multicomponent system, i.e., the so-called Widom–Rowlinson model (cf. Refs. 12–14), which has the remarkable property of displaying a bulk phase transition already in two dimensions, and concentrate on the plane interface in continuous quantum systems (for both bosons and fermions).

2. CONCEPTS, TERMINOLOGY, AND THE STATISTICAL MECHANICAL EXPRESSION OF THE STRESS TENSOR

Since most of the terminology we will rely on has been frequently used in the papers mentioned above (e.g., in Ref. 7 or Ref. 10), only a few remarks are in order here.

We stress the fact that we are exclusively dealing with infinite systems. Then the infinite phase space of classical statistical point mechanics comprises the countable sequences

$$X := \{(x_i),\ x_i := (r_i, p_i) \in \mathbb{R}^d \times \mathbb{R}^d\}$$

It is assumed that the local finiteness property holds, i.e., the set of configurations in phase space with infinitely may particles in a finite volume of coordinate space is of "Gibbs" measure zero. An n-particle observable, say A, is (usually) given by a smooth symmetric function a on $\mathbb{R}^d \times \mathbb{R}^d$ and having a finite support with respect to the position coordinates:

$$A(X) := \sum_{\{i_1, \dots, i_n\}} a(x_{i_1}, \dots, x_{i_n}) \tag{2.1}$$

$\{i_1, \dots, i_n\}$ all ordered n-tuples.

The Poisson bracket of A, B reads

$$\{A, B\}(X) := \sum_i (\partial A/\partial r_i \cdot \partial B/\partial p_i - \partial A/\partial p_i \cdot \partial B/\partial r_i) \tag{2.2}$$

Sometimes slightly more general objects have to be used, such as the particle density:

$$n(r)(X) := \sum_i \delta(r - r_i) \tag{2.3}$$

or the momentum density

$$p(r)(X) := \sum_i p_i \cdot \delta(r - r_i) \tag{2.4}$$

The n-particle distribution functions $\rho^{(n)}(r_1, \dots, r_n)$ are given by

$$\rho^{(n)}(r_1, \dots, r_n) := \sum_{\{i_1, \dots, i_n\}} \langle \delta(r_1 - r_{i_1}) \cdots \delta(r_n - r_{i_n}) \rangle \tag{2.5}$$

with $\langle \cdots \rangle$ the expectation with respect to the thermodynamic equilibrium state. We have in particular

$$\langle n(r) \rangle = \rho^{(1)}(r), \qquad \rho_T^{(2)}(r_1, r_2) = \rho^{(2)}(r_1, r_2) - \rho^{(1)}(r_1) \cdot \rho^{(1)}(r_2) \tag{2.6}$$

with $\rho_T^{(2)}(r_1, r_2) \to 0$ for $|r_1 - r_2| \to \infty$ in a pure phase.

These pure phases of the system are assumed to be prepared by, e.g., the Bogoliubov quasiaverage method or by imposing suitable boundary conditions. We mentioned in the introduction that in case of, e.g., planar interfaces this concept may be problematic, at least for certain systems (preparing a "pure phase" implies in this context that also the phase boundaries have been fixed).

Further concepts will be introduced in the places where they will be needed in Sections 3–5. There is, however, a physical quantity that will be of particular importance in the following and is worthwhile discussing in more detail here. This notion is the statistical mechanical expression for the stress tensor (resp. pressure tensor). We start with its derivation in classical statistical mechanics.

Taking the time derivative of the expression (2.4) for the momentum density $p(r)$, we get (with $H := \sum_i p_i^2/2m + \frac{1}{2}\sum_{i\neq j} V(r_i - r_j)$)

$$\begin{aligned}(d/dt)\, p^\alpha(t) &:= -\{H, p^\alpha(r)\}\\ &= -\partial_{r^\beta}\left[\sum_i (1/m)\, p_i^\alpha p_i^\beta\, \delta(r - r_i)\right]\\ &\quad + \frac{1}{2}\sum_{i\neq j} F_{ij}^\alpha(r_{ij})[\delta(r - r_i) - \delta(r - r_j)]\end{aligned} \tag{2.7}$$

where $r_{ij} := r_i - r_j$, $F_{ij}^\alpha := -\partial V(r_{ij})/\partial r_i^\alpha$, and V is the reflection-invariant two-body interparticle potential (for simplicity we treat only the case of a two-body force). Our strategy is to express the rhs as the divergence of a certain quantity in order to get something like a "conserved current" relation.

The second term on the rhs can be rewritten as

$$\begin{aligned}&\frac{1}{2}\sum_{i\neq j} F_{ij}^\alpha(r_{ij})[\delta(r - r_i) - \delta(r - r_j)]\\ &\quad = \frac{1}{2}\sum_{i\neq j} F_{ij}^\alpha(r_{ij}) \int_0^1 (d/ds)\, \delta(sr_i + (1-s)r_j - r)\, ds\\ &\quad = -\partial_{r^\beta}\left(\frac{1}{2}\sum_{i\neq j} F_{ij}^\alpha(r_{ij}) r_{ij}^\beta \int_0^1 ds\, \delta(sr_i + (1-s)r_j - r)\right)\end{aligned}$$

Equation (2.7) now reads

$$\begin{aligned}(d/dt)\, p^\alpha(r) &= -\partial_{r^\beta}\left[\sum_i (1/m)\, p_i^\alpha p_i^\beta\, \delta(r - r_i)\right.\\ &\quad \left. + \frac{1}{2}\sum F_{ij}^\alpha r_{ij}^\beta \int_0^1 ds\, \delta(sr_i + (1-s)r_j - r)\right]\\ &= \partial_{r^\beta}\sigma^{\alpha\beta}(r)\end{aligned} \tag{2.8}$$

where $-[\cdots]$ is called the statistical mechanical expression for the stress tensor $\sigma^{\alpha\beta}(r)$.

Taking the expectation value of $-\sigma^{\alpha\beta}(r)$, we get the so-called pressure tensor $p^{\alpha\beta}(r)$.

The expression (2.8) was first derived by Irving and Kirkwood[15] by a slightly different method. The nonuniqueness of the definition of the stress tensor is apparent from our derivation. We chose a straight line in the integral $\int_0^1 ds\, \delta(\cdots)$ connecting r_i and r_j. Any other curve joining r_i, r_j

would do a similar job, leading, however, to different $\sigma^{\alpha\beta}(r)$ (in this context cf., e.g., Ref. 16).

In the case of quantum statistics things are slightly more complicated due to the operator character of the expressions. We assume that the particle creation (annihilation) operators $\psi^+(\psi)$ to satisfy Bose (resp. Fermi) commutation relations, i.e.,

$$\begin{gathered}\psi(r)\,\psi^+(r')\mp\psi^+(r')\,\psi(r)=\delta(r-r')\\ \psi^+(r)\,\psi^+(r')\mp\psi^+(r')\,\psi^+(r)=0=\psi(r)\,\psi(r')\mp\psi(r')\,\psi(r)\end{gathered}\tag{2.9}$$

$$\partial_t\psi(r)=(i/\hbar)[H,\psi(r)]\tag{2.10}$$

with

$$H=\frac{\hbar^2}{2m}\int d^dr\,\nabla\psi^+(r)\,\nabla\psi(r)+\frac{1}{2}\iint d^dr\,d^dr'\,\psi^+(r)\,\psi^+(r')\,V(r-r')\,\psi(r')\,\psi(r)$$

The momentum density reads

$$p^\alpha(r)=(\hbar/2i)[\psi^+(r)\,\partial^\alpha\psi(r)-\partial^\alpha\psi^+(r)\,\psi(r)]\tag{2.11}$$

and the counterpart of (2.7) is

$$\begin{aligned}(d/dt)\,p^\alpha(r)= &-(\hbar^2/2m)\,\partial^\beta\{\partial^\beta\psi^+(r)\,\partial^\alpha\psi(r)+\partial^\alpha\psi^+(r)\,\partial^\beta\psi(r)\\ &-\tfrac{1}{2}\partial^\beta\cdot\partial^\alpha[\psi^+(r)\,\psi(r)]\}\\ &-\int\partial_{r^\alpha}V(r-r')\,\psi^+(r)\,\psi^+(r')\,\psi(r')\,\psi(r)\,d^dr'\end{aligned}\tag{2.12}$$

To write this as a divergence of a tensor we again have to transform the second term on the rhs into another form:

$$\begin{aligned}&\int d^dr'\,\partial^\alpha V(r-r')\,\psi^+(r)\,\psi^+(r')\,\psi(r')\,\psi(r)\\ &=\int d^dr'\,d^dr''\,\tfrac{1}{2}(\delta(r-r')-\delta(r-r''))\\ &\quad\times\partial^\alpha V(r'-r'')\,\psi^+(r')\,\psi^+(r'')\,\psi(r'')\,\psi(r')\\ &=-\partial_{r^\beta}\bigg[\int d^dr'\,d^dr''\,\tfrac{1}{2}(r'^\beta-r''^\beta)\,\partial^\alpha V(r'-r'')\\ &\quad\times\int_0^1 ds\,\delta(sr'+(1-s)r''-r)\,\psi^+(r')\,\psi^+(r'')\,\psi(r'')\,\psi(r')\bigg]\end{aligned}\tag{2.13}$$

Thus we arrive at

$$
\begin{aligned}
(d/dt)\, p^{\alpha}(r) = -\partial_{r^{\beta}} \Bigg[& (\hbar^2/2m)\{\partial^{\beta}\psi^{+}(r)\, \partial^{\alpha}\psi(r) + \partial^{\alpha}\psi^{+}(r)\, \partial^{\beta}\psi(r) \\
& - \tfrac{1}{2}\partial^{\alpha}\partial^{\beta}[\psi^{+}(r)\, \psi(r)]\} \\
& - \tfrac{1}{2}\int dr'\, dr''\, (r'^{\beta} - r''^{\beta})\, \partial^{\alpha} V(r' - r'') \\
& \times \int_0^1 ds\, \delta(sr' + (1-s)r'' - r)\, \psi^{+}(r')\, \psi^{+}(r'')\, \psi(r'')\, \psi(r') \Bigg]
\end{aligned}
\tag{2.14}
$$

Again the expression $-[\cdots]$ may be called the microscopic expression for the stress tensor $\sigma^{\alpha\beta}(r)$ in the quantum case.

3. INHOMOGENEOUS CLASSICAL SYSTEMS—GENERAL CASE

We assume that all the inhomogeneities, such as phase boundaries, crystalline or granular structures, etc., have been fixed by appropriate boundary conditions at infinity as indicated in the introduction. While the approach is more or less the same irrespectively of the kind of inhomogeneity, we restrict ourselves for simplicity in this section to inhomogeneities reflected in a nonvanishing density gradient, i.e., $\nabla\rho^{(1)}(r) \neq 0$ at some places (of course, there may be more general types of symmetry breaking, as in liquid crystals).

The connection between a density gradient and higher correlation functions can be established by the expression in the following lemma:

Lemma 1. Let $P_R := \sum_i p_i f_R(r_i)$, where f_R satisfies

(i) $0 \leqslant f_R(r) \leqslant 1$ for all $r \in \mathbb{R}^d$; $f_R \in C^{\infty}(\mathbb{R}^d)$

(ii) $f_R(r) = 1$ for $|r| \leqslant R$

(iii) $f_R(r) = 0$ for $|r| \geqslant R + \varepsilon$

Then the following relation holds:

$$
\nabla\rho^{(1)}(r) = \lim_{R\to\infty} \langle \{P_R, n(r)\} \rangle \tag{3.1}
$$

Proof. Let P_R^α be the αth component of P_R. Then

$$\begin{aligned}\langle\{P_R^\alpha, n(r)\}\rangle &= -\left\langle \sum_j (\nabla_{p_j} P_R^\alpha)[\nabla_{r_j}\delta(r-r_j)]\right\rangle \\ &= -\left\langle \sum_j f_R(r_j)\frac{\partial}{\partial r_j^\alpha}\delta(r-r_j)\right\rangle \\ &= -\int d^dr'\,\rho^{(1)}(r')\,f_R(r')\frac{\partial}{\partial r_j^\alpha}\delta(r-r') \\ &= \frac{\partial}{\partial r^\alpha}[\rho^{(1)}(r)\,f_R(r)] \end{aligned} \tag{3.2}$$

Whenever $R > |r|$, we have $f_R(r) \equiv 1$ and the lemma is proven. ∎

Remark. The reason for making a spatial cutoff in the overall momentum and then taking limits is that $\sum_j p_j$ exists only in a formal sense and is not really a well-defined object (for details see Ref. 9).

Obviously we have

$$\langle\{P_R, n(r)\}\rangle = \langle\{P_R, n(r) - \langle n(r)\rangle\}\rangle \tag{3.3}$$

In the next step we use the classical static KMS condition[11,17]

$$\langle\{A, B\}\rangle = \beta\langle B\{A, H\}\rangle \tag{3.4}$$

yielding in our case

$$\langle\{P_R, n(r)\}\rangle = \beta[\langle n(r)\{P_R, H\}\rangle - \langle n(r)\rangle\langle\{P_R, H\}\rangle] \tag{3.5}$$

While usually the KMS condition has been formulated for bounded observables, it can be shown that it also holds in our more general case. The critical condition is not so much boundedness of observable as their localization in bounded sets of the configuration space (i.e., with respect to the r_i variables).

With $P_R^\alpha = \int d^dr'\, p^\alpha(r')\,f_R(r')$, we can replace $\{P_R^\alpha, H\}$ by $\int d^dr'\, f_R(r')\sum_\beta \partial^\beta\sigma^{\alpha\beta}(r')$ [cf. (2.11)], so that we get

$$\begin{aligned}\partial^\alpha\rho^{(1)}(r) = \lim_{R\to\infty}\beta\int d^dr'\, f_R(r')\Bigg[&\left\langle n(r)\sum_\beta \partial^\beta\sigma^{\alpha\beta}(r')\right\rangle \\ &- \langle n(r)\rangle\left\langle\sum_\beta \partial^\beta\sigma^{\alpha\beta}(r')\right\rangle\Bigg]\end{aligned} \tag{3.6}$$

or, after partial integration,

$$\partial^\alpha \rho^{(1)}(r) = -\beta \lim_{R\to\infty} \int d^d r' \left[\left\langle n(r) \sum_\beta \partial^\beta f_R(r')\, \sigma^{\alpha\beta}(r') \right\rangle - \langle n(r)\rangle \left\langle \sum_\beta \partial^\beta f_R(r')\, \sigma^{\alpha\beta}(r') \right\rangle \right] \tag{3.7}$$

This expression immediately leads to the following theorem about the connection between the presence of inhomogeneities and poor decay of correlations.

Theorem 1. $\nabla\rho^{(1)}(r) = 0$, whenever $\beta < \infty$, and

(i) $\int d^d r\, |r|^\nu |\nabla V(r)| < \infty$ for $0 \leqslant \nu \leqslant 1$

(ii) $\langle n(r)\, \sigma^{\alpha\beta}(r')\rangle - \langle n(r)\rangle \langle \sigma^{\alpha\beta}(r')\rangle = o(|r-r'|^{-(d-1)})$ as $|r-r'| \to \infty$

Proof. Assumption (i) is necessary in order that all expectation values occuring in (3.7) and (ii) are well-defined as we do not assume any further property of the $\rho^{(n)}$ apart from uniform boundedness. Assumption (i) allows us to write (3.7) as

$$\partial^\alpha \rho^{(1)}(r) = -\beta \lim_{R\to\infty} \int d^d r' \sum_\beta \partial^\beta f_R(r') [\langle n(r)\, \sigma^{\alpha\beta}(r')\rangle - \langle n(r)\rangle\langle \sigma^{\alpha\beta}(r')\rangle] \tag{3.8}$$

The support of $\partial^\beta f_R(r')$ is contained in a spherical shell with radii R and $R+\varepsilon$. Therefore, as r is fixed, boundedness of $|\nabla f_R(r)|$ and (ii) imply

$$\int d^d r'\, \partial^\beta f_R(r') [\langle n(r)\, \sigma^{\alpha\beta}(r')\rangle - \langle n(r)\rangle\langle \sigma^{\alpha\beta}(r')\rangle = O(R^{d-1})\, o(R^{-(d-1)}) = o(1) \quad \text{as} \quad R \to \infty$$

Remark 1. Theorem 1 remains valid if $\sigma^{\alpha\beta}(r')$ is replaced by its spherical average

$$\bar\sigma^{\alpha\beta}(r') = \bar\sigma^{\alpha\beta}(|r'|) = \int d\Omega\, \sigma^{\alpha\beta}(|r'|, \Omega)$$

Remark 2. It is one of the merits of employing the notion of stress tensor in this context that one gets space derivatives ∂^β in front of $\sigma^{\alpha\beta}$. By partial integration and $\partial^\beta f_R(r') \equiv 0$ for $|r'| \leqslant R$ it then becomes immediately obvious that $n(r)$ and $\sigma^{\alpha\beta}(r')$ are correlated over long distance if $\partial^\alpha \rho^{(1)}(r) \neq 0$.

Assuming Maxwellian nature of states from now on, we derive some estimates concerning the decay of truncated n-particle density functions. For this purpose, we insert the explicit form of $\sigma^{\alpha\beta}$ into (3.7). Performing some trivial integrations and using

$$\int_0^1 ds\, \delta(sr_1 + (1-s)r_2 - r') = \int_0^1 ds\, \delta((1-s)r_1 + sr_2 - r')$$

we get

$$\begin{aligned}
\partial^\alpha \rho^{(1)}(r) = \beta \lim_{R\to\infty} \sum_\beta \Bigg\{ & \int d^d r' \int d^d p' \\
& \times \partial^\beta f_R(r')\, m^{-1} p'^\alpha p'^\beta [\rho^{(2)}(r, r', p') - \rho^{(1)}(r)\, \rho^{(1)}(r', p')] \\
& + \int d^d p'\, \partial^\beta f_R(r)\, m^{-1} p'^\alpha p'^\beta \rho^{(1)}(r, p') \\
& - \int d^d r' \int d^d r_1 \int d^d r_2\, \partial^\beta f_R(r') \tfrac{1}{2}\, \partial^\alpha V(r_1 - r_2)(r_1^\beta - r_2^\beta) \\
& \times \int_0^1 ds\, \delta(sr_1 + (1-s)r_2 - r')[\rho^{(3)}(r, r_1, r_2) - \rho^{(1)}(r)\, \rho^{(2)}(r_1, r_2)] \\
& - \int d^d r' \int d^d r_1\, \partial^\beta f_R(r')\, \partial^\alpha V(r_1 - r)(r_1^\beta - r^\beta) \\
& \times \int_0^1 ds\, \delta(sr_1 + (1-s)\, r - r')\, \rho^{(2)}(r, r_1) \Bigg\}
\end{aligned} \tag{3.9}$$

The proofs of the following theorems are carried out using various estimates of (3.9):

Theorem 2. $\nabla \rho^{(1)}(r) = 0$, whenever $\beta < \infty$ and

(i) $\int d^d r\, |r|^\nu |\nabla V(r)| < \infty$ for $0 \leqslant \nu \leqslant d$

(ii) $\rho_T^{(2)}(r_1, r_2) = o(|r_1 - r_2|^{-(d-1)})$ as $|r_1 - r_2| \to \infty$

(iii) $\rho_T^{(3)}(r_1, r_2, r_3) = o(|r_1 - r_2|^{-(d-1)})$ uniformly in r_3 as $|r_1 - r_2| \to \infty$

Proof. As we restricted ourselves to Maxwellian states, the first term on the rhs of (3.9) yields

$$\int d^d r' \int d^d p' \sum_\beta \partial^\beta f_R(r')\, m^{-1} p'^\alpha p'^\beta$$
$$\times [\rho^{(2)}(r, r', p') - \rho^{(1)}(r)\, \rho^{(2)}(r', p')]$$
$$= \int d^d r'\, \beta^{-1}\, \partial^\alpha f_R(r')\, \rho_T^{(2)}(r, r')$$
$$= O(R^{d-1})\, o(R^{-(d-1)}) = o(1)$$

as $R \to \infty$. Here we used (ii) and the support property of $\partial^\alpha f_R$, mentioned in the proof of Theorem 1.

The second term on the rhs of (3.9) vanishes whenever $R > |r|$. In order to estimate the third term, we first note that

$$\rho_1^{(3)}(r_1, r_2, r_3) := \rho^{(3)}(r_1, r_2, r_3) - \rho^{(1)}(r_1)\, \rho^{(2)}(r_2, r_3)$$
$$= \rho_T^{(3)}(r_1, r_2, r_3) + \rho^{(1)}(r_2)\, \rho_T^{(2)}(r_1, r_3) + \rho^{(1)}(r_3)\, \rho_T^{(2)}(r_1, r_2) \qquad (3.10)$$

Using (ii) and (iii), we can split this expression into two terms, one of order $o(|r_1 - r_2|^{-(d-1)})$, the other of order $o(|r_1 - r_3|^{-(d-1)})$. Now,

$$\int d^d r' \int d^d r_1 \int d^d r_2\, \partial^\beta f_R(r')\, \partial^\alpha V(r_1 - r_2)(r_1^\beta - r_2^\beta)$$
$$\times \int_0^1 ds\, \delta(s r_1 + (1-s) r_2 - r')\, \rho_1^{(3)}(r, r_1, r_2)$$
$$= \int d^d r' \int d^d r_1 \int d^d r_2'\, \partial^\beta f_R(r')\, \partial^\alpha V(r_1') r_2'^\beta$$
$$\times \int_0^1 ds\, \delta(r_1 - (1-s) r_2' - r')\, \rho_1^{(3)}(r, r_1, r_1 - r_2')$$
$$= \int d^d r' \int d^d r_2'\, \partial^\beta f_R(r')\, \partial^\alpha V(r_2') r_2'^\beta$$
$$\times \int_0^1 ds\, \rho_1^{(3)}(r, r' + (1-s) r_2', r' - s r_2')$$

At this point, it is advantageous to split the integration over r_2' into the two domains, $|r_2'| \leqslant R/2$ and $|r_2'| > R/2$. As the support of $\partial^\beta f_R(r')$ is contained in the shell $R \leqslant |r'| \leqslant R+\varepsilon$, $|r_2'| \leqslant R/2$ implies $|r' + (1-s)r_2'| \geqslant R/2$ and $|r' - sr_2'| \geqslant R/2$ and therefore

$$\rho_1^{(3)}(r, r' + (1-s)r_2', r' - sr_2') = o(R^{-(d-1)})$$

Thus

$$\int d^d r' \int_{|r_2'| \leqslant R/2} d^d r_2' \, \partial^\beta f_R(r') \, \partial^\alpha V(r_2') \times r_2'^\beta \int_0^1 ds \, \rho_1^{(3)}(r, r' + (1-s)r_2', r' - sr_2')$$
$$= O(R^{d-1}) \, o(R^{-(d-1)}) \int_{|r_2'| \leqslant R/2} d^d r_2' \, r_2'^\beta \, \partial^\alpha V(r_2') = o(1)$$

as $|r| \, |\nabla V(r)|$ is integrable.

In case of the other domain $|r_2'| > R/2$ we use uniform boundedness of $|\rho_1^{(3)}|$:

$$\left| \int d^d r' \int_{|r_2'| > R/2} d^d r_2' \, \partial^\beta f_R(r') \, \partial^\alpha V(r_2') r_2'^\beta \times \int_0^1 ds \, \rho_1^{(3)}(r, r' + (1-s)r_2', r' - sr_2') \right|$$
$$\leqslant \int d^d r' \int_{|r_2'| > R/2} d^d r_2' \, |\nabla f_R(r')| \, |\nabla V(r_2')| \, |r_2'| \times \max|\rho_1^{(3)}| \cdot |r_2'|^{d-1} (2/R)^{d-1}$$
$$= O(R^{d-1}) R^{-(d-1)} \int_{|r_2'| > R/2} d^d r_2' \, |r_2'|^d \, |\nabla V(r_2')| = o(1)$$

since the integrability of $|r|^d \, |\nabla V(r)|$ implies

$$\lim_{R \to \infty} \int_{|r| > R/2} d^d r \, |r|^d \, |\nabla V(r)| = 0$$

Provided $R > |r|$, the fourth term on the rhs of (3.9) can be written as follows:

$$\int d^dr' \int d^dr_1 \sum_\beta \partial^\beta f_R(r')\, \partial^\alpha V(r_1 - r)(r_1^\beta - r^\beta)$$
$$\times \int_0^1 ds\, \delta(sr_1 + (1-s)r - r')\, \rho^{(2)}(r, r_1)$$
$$= \int d^dr_1 \sum_\beta \int_0^1 ds\, (r_1^\beta - r^\beta)\, \partial^\beta f_R(sr_1 + (1-s)r)\, \partial^\alpha V(r_1 - r)\, \rho^{(2)}(r, r_1)$$
$$= \int d^dr_1\, [f_R(r_1) - f_R(r)]\, \partial^\alpha V(r_1 - r)\, \rho^{(2)}(r_1, r)$$
$$= \int d^dr_1\, [f_R(r_1) - 1]\, \partial^\alpha V(r_1 - r)\, \rho^{(2)}(r_1, r)$$
$$= \int_{|r_1| > R} d^dr_1\, [f_R(r_1) - 1]\, \partial^\alpha V(r_1 - r)\, \rho^{(2)}(r_1, r)$$

The modulus of this last expression is bounded by

$$\max \rho^{(2)} \int_{|r_1| > R} d^dr_1\, |\nabla V(r_1 - r)|$$

Thus, it vanishes as $R \to \infty$ due to the integrability of $|\nabla V(r)|$.

Theorem 3. $\nabla \rho^{(1)} = 0$, whenever $\beta < \infty$ and

(i) $\int d^dr |\nabla V(r)| < \infty$

(i) $\rho_T^{(2)}(r_1, r_2) = O(|r_1 - r_2|^{-(d+\varepsilon)})$ as $|r_1 - r_2| \to \infty$

(iii) $\rho_T^{(3)}(r_1, r_2, r_3) = O(|r_1 - r_2|^{-(d+\varepsilon)})$ uniformly in r_3 as $|r_1 - r_2| \to \infty$

Proof. The estimates for the first, second, and fourth terms on the rhs of (3.9) given in the proof of Theorem 2 also apply in this case and need not be repeated here.

The third term can be written as follows:

$$\int d^dr' \int d^dr_1 \int d^dr_2 \sum_\beta \partial^\beta f_R(r')\, \partial^\alpha V(r_1 - r_2)(r_1^\beta - r_2^\beta)$$
$$\times \int_0^1 ds\, \delta(sr_1 + (1-s)r_2 - r')\, \rho_1^{(3)}(r, r_1, r_2)$$

$$= \int d^d r_1 \int d^d r_2 \int_0^1 ds \sum_\beta (x_1^\beta - x_2^\beta)\, \partial^\beta f_R(sr_1 + (1-s) r_2)$$
$$\times \partial^\alpha V(r_1 - r_2)\, \rho_1^{(3)}(r, r_1, r_2)$$
$$= \int d^d r_1 \int d^d r_2\, [f_R(r_1) - f_R(r_2)]\, \partial^\alpha V(r_1 - r_2)\, \rho_1^{(3)}(r, r_1, r_2)$$

Conditions (ii), (iii), and (3.10) imply

$$\partial^\alpha V(r_1 - r_2)\, \rho_1^{(3)}(r, r_1, r_2) \in L^1(\mathbb{R}^{2d})$$

for fixed r. Thus, we can use Lebesgue's dominated convergence theorem to write

$$\lim_{R\to\infty} \int d^d r_1 \int d^d r_2\, [f_R(r_1) - f_R(r_2)]\, \partial^\alpha V(r_1 - r_2)\, \rho_1^{(3)}(r, r_1, r_2)$$
$$= \int d^d r_1 \int d^d r_2 \lim_{R\to\infty} [f_R(r_1) - f_R(r_2)]\, \partial^\alpha V(r_1 - r_2)\, \rho_1^{(3)}(r, r_1, r_2)$$
$$= 0 \qquad \text{as} \quad \lim_{R\to\infty} [f_R(r_1) - f_R(r_2)] = 0$$

pointwise in $\mathbb{R}^{2d}$.

Remark 1. Given a non-Maxwellian momentum distribution, Theorems 2 and 3 remain valid under the additional requirement

$$\int d^d p_2\, p_2^\alpha p_2^\beta [\rho^{(2)}(r_1, r_2, p_2) - \rho^{(1)}(r_1)\, \rho^{(1)}(r_2, p_2)]$$
$$= o(|r_1 - r_2|^{-(d-1)})$$

as $|r_1 - r_2| \to \infty$.

Remark 2. One should mention that the statements of Theorems 2 and 3 have already been proved in an approach exploiting the BBGKY hierarchy by Gruber *et al.*[10,21]

It is not astonishing that the use of the KMS condition yields identical results, since classical BBGKY and KMS properties are known to be equivalent in the case of Maxwellian momentum distributions.[11]

We gave an alternative proof for several reasons: First, the techniques employed can immediately be transferred to the quantum case. Second, our proofs seem to throw some additional light on various support and cluster properties as well as the behavior of the kinetic contributions.

Furthermore, Theorems 2 and 3 appear as natural and immediate consequences of our KMS approach which, however, comprises much more, since it also leads to correlations between more general physical observables, such as density, stress, etc.

We conclude this section with the following nice observation:

Proposition 1. Let the KMS state obey strong clustering in time, i.e., $\lim_{t\to\infty}\langle A\cdot B_t\rangle=\langle A\rangle\cdot\langle B\rangle$, with time evolution given by the Liouville operator; then the following holds:

$$\langle A\cdot B\rangle-\langle A\rangle\cdot\langle B\rangle=\beta^{-1}\int_0^{\infty}dt\,\langle\{A,B_t\}\rangle \tag{3.11}$$

Proof.

$$\langle A\cdot B\rangle-\langle A\rangle\cdot\langle B\rangle=\lim_{t\to\infty}\int_t^0 dt'\,(d/dt')\langle A\cdot B_{t'}\rangle=\int_\infty^0 dt'\,\langle A\{B_{t'},H\}\rangle$$

$$=\beta^{-1}\int_0^{\infty}dt\,\langle\{A,B_t\}\rangle$$

(the last equality being a consequence of the KMS condition).

On the one hand, the above relation establishes a link between static (i.e., equal time) and a special sort of dynamic correlation. On the other hand, it is the latter that has a direct analog in the quantum case, while the counterpart of the static correlation does not seem to have any immediate physical interpretation. We shall come back to this point at the end of the next section.

4. INHOMOGENEOUS QUANTUM SYSTEMS—GENERAL CASE

Our approach will be along similar lines as in Section 3 with the proviso that some steps become slightly more complicated due to typical quantum effects. Our methods apply to both bosons and fermions.

Lemma 2. Let $P_R:=\int d^dr\,f_R(r)\,p(r)$ with $p(r)$ of (2.16), $f_R(r)$ as in Lemma 1, and $n(r):=\psi^+(r)\,\psi(r)$. Then

$$\nabla\rho^{(1)}(r)=\nabla\langle n(r)\rangle=\lim_{R\to\infty}\langle(1/i\hbar)[P_R,n(r)]\rangle \tag{4.1}$$

Proof. Relations (2.9) can be used to compute

$$\begin{aligned}(1/i\hbar)[p(r'),\psi^+(r)\,\psi(r)]&=\tfrac{1}{2}[\psi^+(r)\,\delta(r-r')\,\nabla_{r'}\psi(r')\\&\quad-\psi^+(r)\,\nabla_{r'}\delta(r-r')\,\psi(r')-\psi^+(r')\nabla_{r'}\delta(r-r')\,\psi(r)\\&\quad+\nabla_{r'}\psi^+(r')\,\delta(r-r')\,\psi(r)]\end{aligned}$$

Therefore,

$$\langle (1/i\hbar)[P_R, \psi^+(r)\,\psi(r)]\rangle = \left\langle \int d^dr'\, f_R(r')(1/i\hbar)[p(r'), \psi^+(r)\,\psi(r)]\right\rangle$$
$$= \nabla\langle \psi^+(r)\,\psi(r)\, f_R(r)\rangle$$

Whenever $R > |r|$, we have $f_R(r) = 1$ and the lemma is proven. ∎

In the next step we again replace $n(r)$ by $n(r) - \langle n(r)\rangle$ and use the quantum mechanical KMS condition. Denoting time evolution by α_t,

$$\alpha_t A := [\exp(i\tilde{H}t/\hbar)]\, A \exp(-i\tilde{H}t/\hbar)$$

it reads

$$\langle AB\rangle = \langle B\alpha_{i\hbar\beta} A\rangle \tag{4.2}$$

where A and B are elements of the algebra of observables $\mathscr{A}$. As a consequence, we have the relation

$$\langle [A, B]\rangle = \langle B\alpha_{i\hbar\beta} A\rangle - \langle BA\rangle$$
$$= \left\langle \int_0^\beta d\tau\, B\frac{d}{d\tau}(\alpha_{i\hbar\tau} A)\right\rangle = \left\langle \int_0^\beta d\tau\, B\alpha_{i\hbar\tau}[A, \tilde{H}]\right\rangle \tag{4.3}$$

where $\tilde{H}$ denotes the generator of α_t in the concrete representation under discussion. Under certain mild assumptions (e.g., that the time automorphism is approximately inner, i.e., that more or less $\tilde{H} \triangleq \lim_{|\Lambda| \to \infty} (H_\Lambda - H'_\Lambda)$, with H_Λ the formal Hamiltonian of (2.10) for finite volume Λ, assumed to be affiliated with $\mathscr{A}$ (H'_Λ the mirror element being affiliated with its commutant $\mathscr{A}'$), we can replace $[A, \tilde{H}]$ with $[A, H] := \lim_{|\Lambda| \to \infty} [A, H_\Lambda]$ and get

$$\langle [A, B]\rangle = \left\langle \int_0^\beta d\tau\, B\alpha_{i\hbar\tau}[A, H]\right\rangle = \left\langle \int_0^\beta d\tau\, (\alpha_{-i\hbar\tau} B)[A, H]\right\rangle \tag{4.4}$$

the last equality being a consequence of time-translation invariance of the equilibrium state.

Remark. Usually relations like (4.2) and (4.3) are proved for the subclass of analytic elements of $\mathscr{A}$. We assume that time evolution can be extended to $\mathscr{A}$ in the concrete representation under discussion. We even assume the relations to extend to possibly unbounded elements (e.g., products of field operators) which are only affiliated with $\mathscr{A}$ as long as they are sufficiently localized in space (e.g., quasilocal). The possibility of such extensions is discussed in Ref. 22. Nevertheless, the very existence of objects like, e.g., $\psi^+(x)\,\psi(x)$, imaginary time Green's functions, etc., has apparently nowhere been proven for interacting systems and infinite volume. So we have to postulate that expressions like these have a well-defined meaning in some sense (at least after an appropriate renormalization procedure).

In our case, (4.4) yields

$$
\begin{aligned}
\left\langle \frac{1}{i\hbar}\,[P_R^\alpha, n(r)] \right\rangle &= \left\langle \frac{1}{i\hbar}\,[P_R^\alpha, n(r) - \langle n(r)\rangle] \right\rangle \\
&= \left\langle \frac{1}{i\hbar} \int_{-\beta}^{0} d\tau\, [\alpha_{i\hbar\tau} n(r)][P_R^\alpha, H] \right\rangle - \left\langle \int_{-\beta}^{0} d\tau\, \alpha_{i\hbar\tau} n(r) \right\rangle \left\langle \frac{1}{i\hbar}\,[P_R^\alpha, H] \right\rangle \\
&= \int d^d r'\, f_R(r') \left\{ \left\langle \int_{-\beta}^{0} d\tau\, [\alpha_{i\hbar\tau} n(r)] \sum_\beta \partial^\beta \sigma^{\alpha\beta}(r') \right\rangle \right. \\
&\quad \left. - \beta \langle n(r)\rangle \left\langle \sum_\beta \partial^\beta \sigma^{\alpha\beta}(r') \right\rangle \right\} \\
&= - \int d^d r' \left\{ \left\langle \int_{-\beta}^{0} d\tau\, [\alpha_{i\hbar\tau} n(r)] \sum_\beta \partial^\beta f_R(r')\, \sigma^{\alpha\beta}(r') \right\rangle \right. \\
&\quad \left. - \beta \langle n(r)\rangle \left\langle \sum_\beta \partial^\beta f_R(r')\, \sigma^{\alpha\beta}(r') \right\rangle \right\}
\end{aligned} \tag{4.5}
$$

where $\sigma^{\alpha\beta}$ denotes the quantum mechanical stress tensor introduced in (2.14).

Observation. It is known that for a large class of potentials and certain ranges of thermodynamic parameters the so-called reduced density matrices are bounded, continuous functions (cf. Ref. 20 or Corollary 6.3.20 in Ref. 19). Due to the unavoidable lack of normal ordering in certain expressions of (4.5) brought about by the complicated term $\alpha_{i\hbar\tau} n(r)$, we cannot expect this to hold for the expectation values in (4.5). However, on physical grounds we expect the expressions in (4.5) and similar ones below to be bounded continuous functions in r' provided that they are smeared in r with an arbitrary but fixed test function from $\mathscr{D}$ (i.e., a C^∞-function with compact support).

In analogy of Section 3 we get:

Theorem 4. $\nabla\rho^{(1)}(r) = 0$, whenever $\beta < \infty$ and

(i) $\int d^d r\, |r|^\nu\, |\nabla V(r)| < \infty \qquad \text{for} \quad 0 \leqslant \nu \leqslant 1$

$$\text{(ii)} \quad \int d^d r\, h(r) \left\{ \left\langle \int_{-\beta}^{0} d\tau\, [\alpha_{i\hbar\tau} n(r)]\, \sigma^{\alpha\beta}(r') \right\rangle - \left\langle \int_{-\beta}^{0} d\tau\, [\alpha_{i\hbar\tau} n(r)] \right\rangle \langle \sigma^{\alpha\beta}(r') \rangle \right\}$$

$$= o(|r'|^{-(d-1)}) \quad \text{as} \quad |r'| \to \infty \quad \text{for any} \quad h \in \mathscr{D}$$

The proof runs along similar lines as that of Theorem 1. Furthermore, the remarks succeeding the proof of Theorem 1 apply here as well.

Introducing the explicit form of $\sigma^{\alpha\beta}$ into (4.5), we again are able to boil this down to some statements concerning the decay of more elementary correlation functions. However, in contrast to the classical case, it is not the usual truncated n-particle densities that show up, but more complicated density correlations, and, due to the nontriviality of the kinetic part of $\sigma^{\alpha\beta}$, even correlations involving derivatives of field operators. Now, (2.14) and (4.5) yield

$$\begin{aligned}
\partial^\alpha \rho^{(1)}(r) = \lim_{R \to \infty} \sum_\beta \Bigg\{ & \frac{\hbar^2}{2m} \int d^d r\, \partial^\beta f_R(r') \left\langle \int_{-\beta}^{0} d\tau\, [\alpha_{i\hbar\tau} n(r)] \right. \\
& \left. \times [\partial^\alpha \psi^+(r')\, \partial^\beta \psi(r') + \partial^\beta \psi^+(r')\, \partial^\alpha \psi(r')] \right\rangle_T \\
& - \frac{\hbar^2}{4m} \int d^d r'\, \partial^\alpha \partial^\beta \partial^\beta f_R(r') \\
& \times \left\langle \int_{-\beta}^{0} d\tau\, [\alpha_{i\hbar\tau} n(r)][\psi^+(r')\, \psi(r')] \right\rangle_T \\
& - \tfrac{1}{2} \int d^d r' \int d^d r_1 \int d^d r_2\, \partial^\beta f_R(r')(r_1^\beta - r_2^\beta)\, \partial^\alpha V(r_1 - r_2) \\
& \times \int_0^1 ds\, \delta(r' - s r_1 - (1-s) r_2) \Bigg[\left\langle \int_{-\beta}^{0} d\tau\, [\alpha_{i\hbar\tau} n(r)] \right. \\
& \left. \times [\psi^+(r_1)\, \psi^+(r_2)\, \psi(r_2)\, \psi(r_1)] \right\rangle \\
& - \beta \langle n(r) \rangle \langle \psi^+(r_1)\, \psi^+(r_2)\, \psi(r_2)\, \psi(r_1) \rangle \Bigg] \Bigg\}
\end{aligned} \tag{4.6}$$

Here and in the following, $\langle \cdots \rangle_T$ denotes total truncation, defined in the usual way, e.g.: $\langle A(r) \cdot B(r') \rangle_T := \langle A(r) \cdot B(r') \rangle - \langle A(r) \rangle \cdot \langle B(r') \rangle$ etc. (see also (3.10)).

Theorem 5. $\nabla\rho^{(1)}(r)=0$, whenever $\beta<\infty$ and

(i) $\int d^d r\, |r|^\nu |\nabla V(r)|<\infty \quad \text{for} \quad 0\leqslant \nu\leqslant d$

(ii) $$F_h(r'):=\int d^d r\, h(r)\left\langle\left\{\int_{-\beta}^{0} d\tau\, \alpha_{ih\tau}[\psi^+(r)\,\psi(r)]\right\}[\psi^+(r')\,\psi(r')]\right\rangle_T$$
$$=o(|r'|^{-(d-1)}) \quad \text{as} \quad |r'|\to\infty \quad \text{for any} \quad h\in\mathscr{D}$$

(iii) $$G_h(r',r''):=\int d^d r\, h(r)\left\langle\left\{\int_{-\beta}^{0} d\tau\, \alpha_{ih\tau}[\psi^+(r)\,\psi(r)]\right\}\right.$$
$$\left.\times[\psi^+(r')\,\psi^+(r'')\,\psi(r'')\,\psi(r')]\right\rangle_T$$
$$=o(|r'|^{-(d-1)}) \quad \text{uniformly in} \quad r''$$
$$\text{as} \quad |r'|\to\infty \quad \text{for any} \quad h\in\mathscr{D}$$

(iv) $$H_h(r'):=\int d^d r\, h(r)\left\langle\left\{\int_{-\beta}^{0} d\tau\, \alpha_{ih\tau}[\psi^+(r)\,\psi(r)]\right\}\right.$$
$$\left.\times[\partial^\alpha\psi^+(r')\,\partial^\beta\psi^{(r')}+\partial^\beta\psi^+(r')\,\partial^\alpha\psi(r')]\right\rangle_T$$
$$=o(|r'|^{-(d-1)}) \quad \text{as} \quad |r'|\to\infty$$
$$\text{for any} \quad \alpha,\beta \quad \text{and any} \quad h\in\mathscr{D}$$

As the smeared correlations in (ii), (iii), and (iv) are assumed to be bounded functions (see observation above), the proof runs along similar lines as the treatment of the first and third terms on the rhs of (3.9) in the proof of Theorem 2.

Theorem 6. $\nabla\rho^{(1)}=0$, whenever $\beta<\infty$ and

(i) $\int d^d r\, |\nabla V(r)|<\infty$

(ii) $F_h(r')=O(|r'|^{-(d+\varepsilon)})$ as $|r'|\to\infty$ for any $h\in\mathscr{D}$

(iii) $G_h(r',r'')=O(|r'|^{-(d+\varepsilon)})$ uniformly in r'' as $|r'|\to\infty$ for any $h\in\mathscr{D}$

(iv) $H_h(r')=o(|r'|^{-(d-1)})$ as $|r'|\to\infty$ for any $h\in\mathscr{D}$

Again due to the uniform boundedness of F_h, G_h, and H_h, the proof is accomplished by the same strategy as used in those of Theorems 2 and 3.

Remark. The nontriviality of the kinetic contribution is reflected in condition (iv) of the preceding theorems. In classical systems such complications would only arise in case of the (possibly) exotic non-Maxwellian states (cf. remark 1 succeeding the proof of Theorem 3).

As already indicated at the end of the previous section, the at first glance unfamiliar correlation functions F_h, G_h, and H_h, involving imaginary time integrations, can be given physical significance by the following.

Proposition 2. Let the KMS state obey strong clustering in time. Then

$$\int_{-\beta}^{0} d\tau[\langle(\alpha_{i\hbar\tau}A)\cdot B\rangle - \langle A\rangle\langle B\rangle] = \int_0^{\infty} dt\,\langle(1/i\hbar)[A, \alpha_t B]\rangle \tag{4.7}$$

Proof. Using the strong cluster property and (4.3), we get

$$\begin{aligned}
&\int_{-\beta}^{0} d\tau\,[\langle(\alpha_{i\hbar\tau}A)\cdot B\rangle - \langle A\rangle\langle B\rangle] \\
&= \lim_{T\to\infty}\int_T^0 dt\left\langle\int_{-\beta}^{0} d\tau\,(\alpha_{i\hbar\tau}A)(d/dt)(\alpha_t B)\right\rangle \\
&= \int_{\infty}^{0} dt\int_0^{\beta} d\tau\,\langle(\alpha_{-i\hbar\tau}A)\cdot(1/i\hbar)[\alpha_t B, H]\rangle \\
&= \int_0^{\infty} dt\langle(1/i\hbar)[A, \alpha_t B]\rangle \quad \blacksquare
\end{aligned}$$

We also mention Theorem 5.4.12 of Ref. 19 in this context. One sees that the right-hand sides of (3.11) and (4.7) are just the so-called generalized static susceptibilities χ_{AB} of linear response theory.

Thus, we may say that a general feature of inhomogeneous classical and quantum systems is the poor decay of certain static susceptibility functions in position space.

5. THE PLANE INTERFACIAL LAYER, CLASSICAL AND QUANTUM CASES

Since the liquid–gas interface of one-component classical fluids has already been treated in Ref. 7 we will provide only the necessary minimum of concepts and definitions in order to be able to motivate the approach, given in the sequel, for quantum systems.

We study the case of a plane interface layer in d-dimensional space, $r = (x_1, ..., x_d)$. In order also to cover situations like solid–gas or solid–liquid coexistence, we assume the system to be only invariant under the translations belonging to a $(d-1)$-dimensional Bravais lattice with x_d being perpendicular to the $(d-1)$-dimensional hyperplane, $\{r | x_d = 0\}$, containing the lattice. The set of lattice points ρ_i is denoted by G; G_R is a finite piece of G defined by

$$G_R := \{\rho_i, x_1^{(\rho_i)2} + \cdots + x_{d-1}^{(\rho_i)2} \leqslant R^2\} \tag{5.1}$$

Remark. In the case of the spatial coexistence of a crystal and a gas or a liquid one has of course to expect that in the vicinity of the crystal phase boundary there is a certain regrouping of the crystal atoms by which the periodicity of the layers may change. These effects, i.e., a continuous change of periodicity parallel to the surface with the distance from the surface, will be studied elsewhere in order not to overburden the paper with respect to complexity of notation.

Before we come to the quantum case, we would like to make a short aside concerning continuous classical systems consisting of several components. A typical example is the so-called Widom–Rowlinson (WR) model,[12-14] which exhibits a bulk phase transition already in two dimensions and which consists of two species A, B. It can be shown that, after slight modifications, the procedure developed in Ref. 7 can be extended to cover more complex situations like the above with analogous results, e.g., there is no stable A–B interface in two dimensions.

As already remarked in the introduction, there are indications that a flat classical interface cannot be maintained for $d = 3$ in zero gravity. In our view the corresponding arguments are not conclusive in the regime of quantum systems. It is well known that, e.g., in superconductors of the second kind or superfluids interface formation may be enhanced by a possibly negative surface tension (with the orientation of the interfaces being obviously independent of the direction of the gravitational field).

As in Ref. 7, we shall employ from now on a modified cutoff function in the expression for P_R, i.e., we define $f_R(r) = f(|r|/R)$, $f \in C^\infty$, $f(s) = 1$ for $0 \leqslant s < 1$, $f(s) = 0$ for $s > 2$. The main tool in the following will be the quantum version of the Bogoliubov inequality, which, in a first step, can be shown to hold for the subset of analytic elements of the algebra of observables $\mathscr{A}$ and then extended to the full $\mathscr{A}$ by continuity arguments and to unbounded operators that are affiliated with $\mathscr{A}$ under certain technical assumptions (see, e.g., Ref. 22).

Under this proviso the Bogoliubov inequality reads

$$|\langle (i\hbar)^{-1}[A, B]\rangle|^2 \leqslant (\beta/2)\langle B^*B + BB^*\rangle\langle (i\hbar)^{-1}[A, (i\hbar)^{-1}[A^*, H]]\rangle \tag{5.2}$$

We refrain from dropping the factors $(i\hbar)^{-1}$ both to exhibit the correspondence between the classical Poisson bracket $\{A, B\}$ and $(i\hbar)^{-1}[A, B]$ and to be able to identify purely quantum mechanical contributions vanishing in the limit $\hbar \to 0$.

With G, G_R, P_R as above, using (5.2) and the translational invariance under G, we can write for $h \in \mathscr{D}$ and sufficiently large R:

$$\begin{aligned}
\left|\int d^dr\, h(r)\, \partial^\alpha \langle n(r)\rangle\right|^2 &= \left|\int d^dr\, h(r) \frac{1}{|G_R|} \sum_{\rho \in G_R} \partial^\alpha \langle n(r+\rho)\rangle\right|^2 \\
&= \left|\left\langle\left[P_R^\alpha, \int d^dr\, h(r) \frac{1}{|G_R|} \sum_{\rho \in G_R} n(r+\rho)\right]\right\rangle\right|^2 \\
&= \left|\left\langle\left[P_R^\alpha, \int d^dr\, h(r) \frac{1}{|G_R|} \sum_{\rho \in G_R} (n(r+\rho) - \langle n(r)\rangle)\right]\right\rangle\right|^2
\end{aligned} \tag{5.3}$$

$$\begin{aligned}
&\leqslant \beta \int d^dr \int d^dr'\, h(r)\, h(r') \frac{1}{|G_R|^2} \\
&\quad \times \sum_{\rho \in G_R} \sum_{\rho' \in G_R} [\langle n(r+\rho)\, n(r'+\rho')\rangle - \langle n(r)\rangle\langle n(r')\rangle] \\
&\quad \times \left\langle \frac{1}{i\hbar}\left[P_R^\alpha, \frac{1}{i\hbar}[P_R^\alpha, H]\right]\right\rangle \\
&= \beta \int d^dr \int d^dr'\, h(r)\, h(r') \frac{1}{|G_R|^2} \\
&\quad \times \sum_{\rho \in G_R} \sum_{\rho' \in G_R} [\langle \psi^+(r+\rho)\, \psi^+(r'+\rho')\, \psi(r'+\rho')\, \psi(r+\rho)\rangle_T \\
&\quad + \delta(r+\rho-r'-\rho')\langle \psi^+(r+\rho)\, \psi(r+\rho)\rangle] \left\langle \frac{1}{i\hbar}\left[P_R^\alpha, \frac{1}{i\hbar}[P_R^\alpha, H]\right]\right\rangle \\
&= \beta \left\langle \frac{1}{i\hbar}\left[P_R^\alpha, \frac{1}{i\hbar}[P_R^\alpha, H]\right]\right\rangle \sum_{\rho \in G_R} \sum_{\rho' \in G_R} \frac{1}{|G_R|^2}\left[\int d^dr \int d^dr'\, h(r)\, h(r')\right. \\
&\quad \left. \times \rho_T^{(2)}(r+\rho, r'+\rho') + \int d^dr\, h(r)\, h(r+\rho-\rho')\, \rho^{(1)}(r)\right]
\end{aligned} \tag{5.4}$$

The double sum in this last expression is identical to that arising in the classical case. (Note that in the corresponding equation of Ref. 7 a term is missing.) Therefore for $d \geqslant 2$ we have

$$\sum_{\rho \in G_R} \sum_{\rho' \in G_R} \frac{1}{|G_R|^2}(\cdots) = o(R^{-(d-2)}) \qquad \text{as} \quad R \to \infty$$

whenever $\rho_T^{(2)}(r, r') = o(|r-r'|^{-(d-2)})$ as $|r-r'| \to \infty$. For $d=1$ there is no lattice average and the double sum reduces to

$$\int dr \int dr'\, h(r)\, h(r')\, \rho_T^{(2)}(r, r') + \int dr\, h^2(r)\, \rho^{(1)}(r)$$

which is bounded and independent of R. Thus, whenever we are able to show that

$$\left\langle \frac{1}{i\hbar}\left[P_R^\alpha, \frac{1}{i\hbar}\,[P_R^\alpha, H]\right]\right\rangle = O(R^{d-2}) \qquad \text{for any} \quad d \geqslant 1$$

by (5.4) we can conclude that $\nabla\rho^{(1)}(r) = 0$. The circumstances under which this is possible are stated in the following:

Theorem 7. Let $\beta < \infty$. Then $\nabla\rho^{(1)}(r) \equiv 0$ if the following holds:

(i) $\rho^{(1)}(r)$, $\rho^{(2)}(r, r')$, $\langle \partial^\alpha\psi^+(r)\,\partial^\beta\psi(r) + \partial^\beta\psi^+(r)\,\partial^\alpha\psi(r)\rangle$ are bounded functions,

(ii) $\rho_T^{(2)}(r, r') = o(|r-r'|^{-(d-2)})$ as $|r-r'| \to \infty$

(iii) $\int d^dr\, |r|^2\, |(\partial^\alpha)^2\, V(r)| < \infty, \quad 1 \leqslant \alpha \leqslant d$

(iv) $\rho^{(1)}(r) = \rho^{(1)}(r+\rho)$; $\rho^{(2)}(r, r') = \rho^{(2)}(r+\rho, r'+\rho)$ for all $\rho \in G$

Proof. The calculation of the twofold commutator $(1/i\hbar)[P_R^\alpha, (1/i\hbar)[P_R^\alpha, H]]$ is somewhat lengthy and cumbersome, but nothing but a straightforward application of the (anti-) commutation relations (2.9). We quote the result:

$$\begin{aligned}
&\frac{1}{i\hbar}\left[P_R^\alpha, \frac{1}{i\hbar}\,[P_R^\alpha, H]\right] \\
&\quad = \frac{\hbar^2}{4m}\int d^dr \sum_\beta \partial^\beta f_R(r) \\
&\qquad \times \{2f_R(r)\,\partial^\alpha[\partial^\alpha\psi^+(r)\,\partial^\beta\psi(r) + \partial^\beta\psi^+(r)\,\partial^\alpha\psi(r)] \\
&\qquad + 4\partial^\beta f_R(r)\,\partial^\alpha\psi^+(r)\,\partial^\alpha\psi(r) \\
&\qquad + 3\partial^\alpha f_R(r)[\partial^\alpha\psi^+(r)\,\partial^\beta\psi(r) + \partial^\beta\psi^+(r)\,\partial^\alpha\psi(r)] \\
&\qquad - \partial^\alpha\partial^\beta f_R(r)\,\partial^\alpha[\psi^+(r)\,\psi(r)] - \partial^\alpha\partial^\alpha\partial^\beta f_R(r)\,\psi^+(r)\,\psi(r)
\end{aligned}$$

$$- \partial^\beta f_R(r)\, \partial^\alpha\partial^\alpha[\psi^+(r)\,\psi(r)] - f_R(r)\, \partial^\alpha\partial^\alpha\partial^\beta[\psi^+(r)\,\psi(r)]$$
$$- 2\partial^\alpha f_R(r)\, \partial^\alpha\partial^\beta[\psi^+(r)\,\psi(r)]\}$$
$$+ \tfrac{1}{2}\int d^d r \int d^d r'\, [f_R(r) - f_R(r')]^2$$
$$\times (\partial^\alpha)^2\, V(r-r')\, \psi^+(r)\, \psi^+(r')\, \psi(r')\, \psi(r)$$
$$+ \int d^d r \int d^d r'\, f_R(r)\, \partial^\alpha f_R(r)\, \partial^\alpha V(r-r')$$
$$\times \psi^+(r)\, \psi^+(r')\, \psi(r')\, \psi(r) \tag{5.5}$$

The last term in this expression is the only one that does not seem to behave well enough in the limit $R \to \infty$. But since we are only interested in expectation values, it can be canceled by the following trick, well known from the classical case.[7] Define

$$\tilde{P}_R^\alpha := \int d^d r\, f_R(r)\, \partial^\alpha f_R(r)\, p^\alpha(r)$$

By time translation invariance of KMS states we have $\langle (1/i\hbar)[\tilde{P}_R^\alpha, H]\rangle = 0$ and therefore

$$\left\langle \frac{1}{i\hbar}\left[P_R^\alpha, \frac{1}{i\hbar}[P_R^\alpha, H]\right]\right\rangle = \left\langle \frac{1}{i\hbar}\left[P_R^\alpha, \frac{1}{i\hbar}[P_R^\alpha, H]\right] + \frac{1}{i\hbar}[\tilde{P}_R^\alpha, H]\right\rangle \tag{5.6}$$

The calculation of $(1/i\hbar)[\tilde{P}_R^\alpha, H]$ is again straightforward and yields

$$\frac{1}{i\hbar}[\tilde{P}_R^\alpha, H] = \frac{\hbar^2}{4m}\int d^d r \sum_\beta f_R(r)\, \partial^\beta f_R(r)$$
$$\times \{\partial^\alpha\partial^\alpha\partial^\beta[\psi^+(r)\,\psi(r)] - 2\partial^\alpha[\partial^\alpha\psi^+(r)\,\partial^\beta\psi(r) + \partial^\beta\psi^+(r)\,\partial^\alpha\psi(r)]\}$$
$$- \int d^d r \int d^d r'\, f_R(r)\, \partial^\alpha f_R(r)\, \partial^\alpha V(r-r')\, \psi^+(r)\, \psi^+(r')\, \psi(r')\, \psi(r)$$

Thus, using (5.6) and performing some partial integrations, we have the final result:

$$\left\langle \frac{1}{i\hbar}\left[P_R^\alpha, \frac{1}{i\hbar}[P_R^\alpha, H]\right]\right\rangle$$
$$= \frac{\hbar^2}{m}\int d^d r \sum_\beta \partial^\beta f_R(r)\, \partial^\beta f_R(r) \langle \partial^\alpha\psi^+(r)\, \partial^\alpha\psi(r)\rangle$$
$$+ \frac{3\hbar^2}{4m}\int d^d r \sum_\beta \partial^\alpha f_R(r)\, \partial^\beta f_R(r)\, \langle \partial^\alpha\psi^+(r)\, \partial^\beta\psi(r) + \partial^\beta\psi^+(r)\, \partial^\alpha\psi(r)\rangle$$

$$-\frac{\hbar^2}{4m}\int d^dr \sum_\beta \{4\partial^\alpha\partial^\alpha\partial^\beta f_R(r)\,\partial^\beta f_R(r)+2\partial^\alpha\partial^\beta\partial^\beta f_R(r)\,\partial^\alpha f_R(r)$$
$$+\,3\partial^\alpha\partial^\beta f_R(r)\,\partial^\alpha\partial^\beta f_R(r)+2\partial^\alpha\partial^\alpha f_R(r)\,\partial^\beta\partial^\beta f_R(r)\}\,\rho^{(1)}(r)$$
$$+\,\tfrac{1}{2}\int d^dr\int d^dr'\,[f_R(r)-f_R(r')]^2(\partial^\alpha)^2\,V(r-r')\,\rho^{(2)}(r,r') \qquad (5.7)$$

With the explicit form $f_R(r)=f(|r|/R)$, we see that $\partial^\alpha f_R(r)=O(R^{-1})$ as $R\to\infty$; thus, the first two terms on the rhs of (5.7) are clearly $O(R^{d-2})$.

The fourth term is identical to the one appearing in the classical case and is therefore also of order R^{d-2}.[7]

As to the third term, we note that with our choice of f_R, contrary to a surmise of Martin,[23] the higher derivatives of f_R do not cause any trouble whatsoever, but have an even faster decay. The expression in curly brackets is immediately seen to be $O(R^{-4})$; thus the third term is of order R^{d-4} and therefore completely well-behaved.

Remark 1. The assumed boundedness of $\langle\partial^\alpha\psi^+\partial^\beta\psi+\partial^\beta\psi^+\partial^\alpha\psi\rangle$ is connected with the rather natural requirement that the pressure tensor should be a bounded quantity.

Remark 2. The third term on the rhs of (5.7) is easily identified as a purely quantum mechanical contribution vanishing in the limit $\hbar\to 0$. This term is $O(R^{d-4})$ and thus reflects the harmlessness of quantum corrections like these.

Remark 3. Theorem 7 remains valid if the clustering assumption

$$\rho_T^{(2)}(r,r')=o(|r-r'|^{-(d-2)}) \qquad \text{as} \quad |r-r'|\to\infty$$

is replaced by the weak clustering assumption

$$\frac{1}{|G_R|}\sum_{\rho\in G_R}\rho_T^{(2)}(r,r'+\rho)=o(R^{-(d-2)}) \qquad \text{as} \quad R\to\infty$$

Remark 4. Examples covered by our procedure are plane liquid–gas or liquid–crystal quantum interfaces as well as purely crystalline states.

Remark 5. Under the assumptions made in Theorem 7 or remark 3, $\nabla\rho^{(1)}\equiv 0$, whenever $d\leqslant 2$, provided that there is some sort of clustering of $\rho_T^{(2)}$ at all.

REFERENCES

1. J. S. Rowlinson and B. Widom, *Molecular Theory of Capillarity* (Oxford University Press, 1982).
2. J. K. Percus, Non-uniform fluids, in *The Liquid State of Matter*, E. W. Montroll and J. L. Lebowitz, eds. (North-Holland, 1982).
3. C. A. Croxton, *Statistical Mechanics of the Liquid Surface* (Wiley, New York, 1980).
4. R. B. Griffith and D. E. Wolf, Preprint, Carnegie-Mellon University, Pittsburgh (1985).
5. F. P. Buff, R. A. Lovett, and F. H. Stillinger, *Phys. Rev. Lett.* **15**:621 (1965); M. S. Wertheim, *J. Chem. Phys.* **65**:2377 (1976); J. D. Weeks, *J. Chem. Phys.* **67**:3106 (1977).
6. J. K. Percus, *Faraday Symp. Chem. Soc.* **16** (1981).
7. M. Requardt, *J. Stat. Phys.* **31**:679 (1983).
8. M. Robert, *Phys. Rev. Lett.* **54**:444 (1985).
9. M. Requardt, *Z. Phys. B* **36**:187 (1979).
10. C. Gruber, P. A. Martin, and C. Oguey, *Commun. Math. Phys.* **84**:55 (1982).
11. G. Gallavotti and E. Verboven, *Nuovo Cimento* **28B**:274 (1975).
12. B. Widom and J. S. Rowlinson, *J. Chem. Phys.* **52**:1670 (1970).
13. D. Ruelle, *Phys. Rev. Lett.* **27**:1040 (1971).
14. J. L. Lebowitz and E. Lieb, *Phys. Lett.* **39A**:98 (1972).
15. J. H. Irving and J. G. Kirkwood, *J. Chem. Phys.* **18**:817 (1950).
16. P. Schofield and J. R. Henderson, *Proc. R. Soc. Lond. A* **379**:231 (1982).
17. N. D. Mermin, *J. Math. Phys.* **8**:1061 (1967).
18. J. G. Kirkwood and F. P. Buff, *J. Chem. Phys.* **17**:338 (1949).
19. O. Bratteli and D. W. Robinson, *Operator Algebras and Quantum Statistical Mechanics* II, (Springer, New York, 1981).
20. J. Ginibre, *J. Math. Phys.* **6**:238 (1965).
21. C. Gruber and P. A. Martin, *Ann. Phys.* **131**:56 (1981).
22. J. G. Garrison and J. Wong, *Commun. Math. Phys.* **26**:1 (1972).
23. P. A. Martin, *Nuovo Cimento* **68B**:302 (1982).