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Uniform Upper Bounds (and Thermodynamic Limit) for the Correlation Functions of Symmetric Coulomb-Type Systems

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Uniform upper bounds are proven for the correlation functions in the strictly charge-neutral canonical and grand canonical ensembles for charge-symmetric two-component systems. For the grand canonical ensemble the increase of the correlation functions along the thermodynamic-limit sequence is shown as well, implying the existence of the states. The particles have bounded pair interactions of positive type. Both classical and quantum systems with Boltzmann statistics are considered. Coulomb systems with regularized interactions are included as a special case.

KEY WORDS: Coulomb systems; correlation functions; thermodynamic limit for states; Siegert transformation; Gaussian functional integrals; Wiener measure.

1. INTRODUCTION

The existence of the thermodynamic limit (T-limit) for the thermodynamic functions of Coulomb systems is a well-known result due to Lieb and Lebowitz.[1] The existence of the thermodynamic states is a problem that has only partially been solved. Eventually one would like to prove the existence of states in the various statistical ensembles for the physical matter system, which is a quantum mechanical system of negatively charged fermions (electrons) and several species of positively charged particles which may be either fermions or bosons. However, we seem to be far away from such a goal.

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So far, all results about the existence of states of Coulomb systems pertain to the unrestricted grand canonical ensemble (GCE), with the activities adjusted so that the mean charge is zero. We will call this ensemble the mean neutral GCE. Existence of the correlation functions at all temperatures and activities, in both classical and quantum statistical mechanics, has been established for charge-conjugation-invariant (symmetric) systems with bounded interactions of positive type by Fröhlich and Park.[2,3] For nonsymmetric classical systems, existence has so far been shown only for the high-temperature, low-density regime; in that case also the exponential clustering of the correlation functions was proved. These results are due to Brydges and Federbush.[4] Imbrie[5] gave a corresponding proof for jellium and inquired into the screening of fractional charges.

The objective of the present paper is to proceed a small step further in the direction of extending the above-mentioned results to the other ensembles. More precisely, it will be shown that the results of Fröhlich and Park[2] can be extended to the strictly neutral grand canonical ensemble, and at least part of that analysis to the (strictly) neutral canonical ensemble (CE) for charge-conjugation-invariant systems. In the strictly neutral GCE, the system is constrained to contain an equal number of positive and negative charges. In contrast, the mean-neutral unrestricted GCE allows for charge fluctuations about the strictly neutral case. A neutral CE, on the other hand, is automatically strictly neutral. The particles interact with continuous potentials of positive type. Coulomb systems with regularized interactions are included as a special case. Only the short-range regularization is required. There are no restrictions that come from the long-range character of the Coulomb interactions.

To prove the existence of the states, one has to control the correlation functions along the T-limit sequence. For the mean-neutral unrestricted GCE for symmetric systems, Fröhlich and Park proved uniform upper bounds on the correlation functions, and also the increase of the correlation functions along the T-limit sequence. Together these results imply the existence of the T-limit for the correlation functions, hence for the states. Their method was to use the Siegert representation[6] and Ginibre's[7] technique of proving correlation inequalities.

The Siegert representation is a representation of the correlation functions with the help of Gaussian functional integrals, combined with the Feynman–Kac formula in the quantum case. A predecessor of it is found in a paper by Stratonovich,[8] who worked with the second quantization formalism. In this representation some estimates are readily done which are outside of sight or at least obscured in the standard representation. In particular, the classical correlation functions of charge-conjugation-invariant systems in the mean-neutral unrestricted GCE can be written as an expectation functional in the form

$$\varrho_n(\mathbf{r}_1,..., \mathbf{r}_n) = \left\langle \prod_{k=1}^{n} ce^{i\sigma_k \phi(\mathbf{r}_k)} \right\rangle$$

where $c > 0$ is a constant (with respect to volume and the Gaussian field variable ϕ), $\langle \cdot \rangle$ is an average, and $\sigma_k = \pm 1$. The uniform (with respect to the volume) upper bounds $\varrho_n \leqslant c^n$ are immediately obtained from the elementary inequality $|\langle F \rangle| \leqslant \langle |F| \rangle$, since here $|F| = \sup |F| = c^n$.

To prove the increase of the correlation functions, Fröhlich and Park made use of the following fact. In the mean-neutral unrestricted GCE the expectation functional $\langle \cdot \rangle$ has the structure of a formal thermal average with formal Hamiltonian of a continuum analog of the plane rotator model. This allows one to apply Ginibre's[7] general formalism for proving correlation inequalities. For the discussion given below it is useful to mention here that the relevant inequality in refs. 2 and 3 is of the type

$$\langle \cos \phi(\mathbf{r}) \cos \phi(\mathbf{r}') \rangle - \langle \cos \phi(\mathbf{r}) \rangle \langle \cos \phi(\mathbf{r}') \rangle \geqslant 0 \tag{1.1}$$

It can be proved using duplicate variables and orthogonal transformations in field space. Using (1.1), the monotonic increase of the correlation functions along the T-limit sequence can be established by an interpolation argument.[2] The quantum version with Boltzmann statistics[2] has essentially the same features, except that $ce^{\pm i\phi}$ has to be replaced by a Wiener integral (see also the Appendix).

In this paper we show that the above analysis can be extended to the strictly neutral GCE and partly to the neutral CE. More precisely, we prove uniform upper bounds with respect to the volume and the total number of particles for the correlation functions in the neutral canonical ensemble for symmetric two-component systems. Corresponding bounds, with respect to the volume, for the correlation functions in the strictly neutral grand canonical ensemble are constructed as well. In a second step we can prove increase of the grand canonical correlation functions along the sequence, which together with the bounds allows us to conclude the existence of limits for the T-limit sequence of the correlation functions of the GCE.

The Siegert representation will play a decisive role in the analysis. However, the techniques that have been used[2] for the mean-neutral unrestricted GCE need to be modified, to the extent that for parts of the program the Siegert representation has to be transformed further.

To estimate the upper bounds, the Siegert representation is appropriate, but instead of essentially a single inequality,[2] a sequence of inequalities is needed for the strictly neutral CE and GCE. It will also be shown that the special cases of the neutral correlation functions of the strictly neutral GCE are estimated from above in essentially the same way as done by Fröhlich and Park.

The results of Fröhlich and Park already show that the second part of the program, i.e., monotonicity along the T-limit sequence, is harder to prove. Indeed, if one attempts to adapt the techniques of Fröhlich and Park to the Siegert representation of the strictly neutral CE and GCE, one encounters new difficulties. In particular, the Siegert representation of these ensembles does not have the structure of a formal thermal average of plane rotator systems, but introduces structures which seem to require qualitatively new arguments to prove the existence of states. This problem can be overcome for the strictly neutral GCE. One can employ a second averaging process which in combination with the Siegert representation transforms the strictly neutral GCE into a form essentially identical to the "bare" Siegert representation of the unrestricted mean neutral GCE. The techniques of Fröhlich and Park can be adapted and the increase of the correlation functions shown. A minor modification is that we will avoid the interpolation argument of ref. 2.

In this averaged Siegert representation, the bounds for all the grand canonical correlation functions can now be obtained as in ref. 2, alternatively. Nevertheless, the greater ease with which the bounds can now be obtained, as compared to the bare Siegert representation, is compensated by the additional effort of introducing the additional averaging.

Unfortunately, so far it has proven elusive to establish the increase of the canonical correlation functions. The relevant inequalities that are needed to prove the increase are no longer of the type (1.1). On the other hand, they are also not too different, and it is possible to use the basic technical ingredients of Ginibre's formalism to manipulate the expressions, i.e., duplicate variables and orthogonal transformations in field space. Unfortunately the resulting expressions which are expected to be positive are not manifestly so. The author's opinion is that this is merely a technical difficulty and that one will eventually be able to show the increase. Since the calculations have not yet been conclusive they will not be included in the present paper, however. Instead, in Appendix B we supplement the present analysis by a new proof of the subadditivity of the canonical free energy (recovering a result due to Griffiths[9]) as well as strong superadditivity of the grand canonical potential (a new result). Strong sub(super)additivity of thermodynamic potentials is interesting for its own sake, but also since the technical difficulties are essentially the same as for proving increase of the correlation functions.

The results obtained here still pertain to symmetric systems which are neutral at least in the mean. For other Coulomb-type systems the situation is more complicated. It can be inferred from the work of Lieb and Lebowitz[1] that a small amount of net charge should be allowed in the system, but the T-limit then shows a capacity effect and becomes shape-dependent. This introduces additional complications even for symmetric systems. For the physical matter system there are charges of different magnitudes and sign, and charge-conjugation invariance is lacking. An idea of what the problems might be in that case is mediated by a comparison between the proofs of the thermodynamic functions for the charge-symmetric systems[9] (see also refs. 2 and 3) and for the matter system.[1]

Last, and not least, it should be mentioned that the upper bounds proved here and their counterparts proved in ref. 2 have potential further applications. They are useful for estimating certain expectation values of the physical observables. An application to the screening problem of Coulomb systems at high densities will be given in a subsequent work.

2. BASIC SETUP

The discussion will not be restricted to Coulomb systems with regularized interactions, but these are the prominent examples of the kind of system considered here. Therefore, in a convenient abuse of language, one may speak of positively and negatively "charged" particles also for the systems with non-Coulomb interactions. A system consists of two species of particles of equal and opposite charges. There is an equal number of positive and negative particles in a system. If $V_{k,j}(|\mathbf{r}_k - \mathbf{r}_j|)$ is the pair interaction energy between two particles at positions $\mathbf{r}_k$ and $\mathbf{r}_j$, then $V_{k,j} = +V$ if the two particles belong to the same species, and $V_{k,j} = -V$ otherwise. The function $V\colon \mathbb{R}^+ \cup \{0\} \to \mathbb{R}$ has strictly positive Fourier transform. Such a function is said to be of positive type, for it defines the kernel of a positive bilinear form on some Hilbert space. In order to assure thermodynamic stability[10] for all temperatures,[3] a further requirement is that the potential V is bounded above by $V_0 = V(0) < \infty$. We require V to be continuous, so that boundedness is automatically given.

The Hamiltonian of a finite system is

$$H^{(N)} = K^{(N)} + U^{(N)} \tag{2.1}$$

[3] If one drops the requirement that thermodynamic stability should hold for all temperatures, then V need not be bounded. An example is a classical Coulomb system in two space dimensions. Although the logarithmic singularity of the interactions does not allow for an extensive lower bound on the total potential energy of a microscopic configuration, the T-limit exists for high enough temperatures.[11,12]

where

$$K^{(N)} = \sum_{j=1}^{2N} (2m_j)^{-1} |\mathbf{p}_j|^2 \tag{2.2}$$

is the kinetic energy and

$$U^{(N)} = \sum_{1 \leqslant k < j \leqslant 2N} V_{k,j}(|\mathbf{r}_k - \mathbf{r}_j|) \tag{2.3}$$

the total potential energy, in standard notation. The potential energy satisfies the stability requirement $U \geqslant -BN$, with $B = V_0$, shown by Fisher and Ruelle.[10] This implies quantum stability as well.[10] That way already the classical Hamiltonian mimicks stabilizing quantum effects, although it is clearly only a crude approximation to more realistic fermion systems.

For the classical case the finite system's canonical correlation functions on the phase space $\mathbb{R}^{6N} \times \Lambda^{2N}$, with $\Lambda \subset \mathbb{R}^3$ bounded, factorize into a momentum and a configuration-space part. The momentum part of the total correlation function is trivial, and this is true for the grand canonical ensemble as well. The objects of interest in the present paper are the configuration-space correlation functions for the canonical and grand canonical ensembles.[4] They are conveniently obtained from the marginal densities of the canonical probability density of a finite system. The canonical configurational probability density (with respect to normalized Lebesgue measure; see below) is given by

$$\eta^{(N,\Lambda)}(\mathbf{r}_1,\dots,\mathbf{r}_{2N}) = N!^{-2} Z^{-1} \exp(-\beta U^{(N)}) \tag{2.4a}$$

where

$$Z(N, \Lambda) = N!^{-2} \int_{\Lambda^{2N}} \exp(-\beta U^{(N)}) \prod_{l=1}^{2N} \kappa(d^3 r_l) \tag{2.4b}$$

is the classical canonical partition function. The quantization of phase space is taken into account in the usual heuristic manner. In that sense, $\kappa(d^3 r) = \lambda_{\mathrm{dB}}^{-3} d^3 r$ is the (heuristically) normalized Lebesgue measure, with $\lambda_{\mathrm{dB}} = (h^2 \beta / 2\pi \bar{m})^{1/2}$ the thermal de Broglie wavelength. We denote by $\bar{m}$ the geometric mean of m_+ and m_-. Let us stipulate that in the further discussion $m_+ = m_- = m$, for in the quantum case we have to postulate this for technical reasons.[9] In the classical case the generalization to $m_+ \neq m_-$ is straightforward. Explicit mention of β as a variable of Z has been omitted,

[4] Since no nonneutral systems will be considered, the attribute "strictly neutral" will be dropped from now on except in case we need to distinguish between the two GCEs.

since β plays a trivial role in the considerations of the present paper. A marginal probability density $\eta^{(N,\Lambda)}_{n_+,n_-}$ for n_+ positive and n_- negative particles, with $n_\pm \leqslant N$, is obtained by integrating (2.4a) with the normalized Lebesgue measure over all $2N - n_+ - n_-$ remaining variables. Instead of writing down here the explicit expression in the standard representation given by (2.4), a more compact form will be established in the next section.

The canonical configurational correlation function $\rho^{(N,\Lambda)}_{n_+,n_-}$ is given by

$$\rho^{(N,\Lambda)}_{n_+,n_-} = c(N; n_+, n_-)\, \eta^{(N,\Lambda)}_{n_+,n_-} \tag{2.5a}$$

with

$$c(N; n_+, n_-) = \prod_{j=0}^{\hat{j}} \prod_{k=0}^{\hat{k}} (N-j)(N-k) \tag{2.5b}$$

and with $\hat{j} = n_+ - 1$; $\hat{k} = n_- - 1$. The convention $\prod_{k=0}^{-1}(N-k) = 1$ is to be employed if either n_+ or n_- vanishes. For the GCE the corresponding correlation function $\varrho^{(\Lambda)}_{n_+,n_-}$ is (with $\check{n} = \max\{n_+, n_-\}$)

$$\varrho^{(\Lambda)}_{n_+,n_-} = \Xi^{-1} \sum_{N=\check{n}}^{\infty} z^{2N} Z(N, \Lambda)\, \rho^{(N,\Lambda)}_{n_+,n_-} \tag{2.6a}$$

where

$$\Xi(\Lambda) = \sum_{N=0}^{\infty} z^{2N} Z(N, \Lambda) \tag{2.6b}$$

is the classical grand canonical partition function, and $z = \exp(\beta\mu)$, with μ the chemical potential.

For the quantum mechanical systems the Hamiltonian (2.1) has to be interpreted as an operator. The particle density correlation functions are obtained from the configuration-space representation of the reduced density matrices. This means that $H^{(N)}$ is acting on $\bigotimes_{j=1}^{2n} L^2(\Lambda, d^3r_j)$, with $\mathbf{p}_j^2$ replaced by $-\hbar^2\nabla_j^2$. The classical configuration-space integrals have analogs in the quantum mechanical setting which are obtained from the configuration-space representation of $\mathrm{Tr}\, e^{-\beta H^{(N)}}$. Gaussian integrals have to be combined with Wiener integrals.[6] The quantum mechanical version with Boltzmann statistics[2,6] is essentially a straightforward generalization of the classical one, although somewhat more complicated in the basic setting. However, the actual expected benefits of a quantum mechanical formulation of the Coulomb problem, namely the possibility of treating the exact Coulomb potential for a multispecies system that is not charge-conjugation invariant, if only one of the species consists of fermions, do not appear by this method. Thus, one does not gain the desired insight into the physical matter problem[1] by a quantum version of charge-symmetric systems with Boltzmann statistics. For that reason the quantum version, which is included for the sake of completeness, is only summarized in an appendix to this paper.

3. GAUSSIAN FUNCTIONAL INTEGRAL REPRESENTATION OF THE CLASSICAL CANONICAL ENSEMBLE

For convenience, the representation of the density of the canonical equilibrium measure in terms of Gaussian function space integrals[6] is recalled here. The representation of the other ensembles then follows. For a general discussion of Gaussian integrals on function space, and also for the Hilbert spaces $\mathscr{H}_{-\infty}$ and $\mathscr{H}_{\infty}$ which are mentioned below, see, e.g., Appendix A of the book by Glimm and Jaffe.[13] Here it may suffice to think of $\mathscr{H}_{\infty}$ as the C^{∞} functions of rapid decrease at infinity, equipped with a Hilbert space structure, and $\mathscr{H}_{-\infty}$ as the space of tempered distributions. For further applications and discussion see also the early contributions by Albeverio and Høegh-Krohn,[14] Edwards and Lenard,[15] and Stratonovich,[8] as well as Simon's book.[16]

To rewrite $\exp(-\beta U)$ in terms of Gaussian integrals, for $f \in \mathscr{H}_{\infty}$ let $d\gamma(\Phi)$ be the Gaussian measure on $\mathscr{H}_{-\infty}$ with covariance βV and mean 0, such that

$$\int e^{i\Phi(f)}\, d\gamma(\Phi) = e^{-(1/2)\langle f, f\rangle} \tag{3.1}$$

with

$$\int d\gamma(\Phi) = 1 \tag{3.2}$$

where $\langle \cdot, \cdot \rangle$ is a positive-definite bilinear form on $\mathscr{H}_{\infty} \times \mathscr{H}_{\infty}$ with kernel βV (Minlos' Theorem). Averages[5] taken with that measure will be abbreviated by angular brackets,

$$\int \cdot\, d\gamma(\Phi) = \langle \cdot \rangle \tag{3.3}$$

Wick ordering is defined by

$$:e^{i\Phi(f)}: = e^{(1/2)\langle f, f\rangle} e^{i\Phi(f)} \tag{3.4}$$

[5] There should be no confusion with the usual statistical averages. Angular brackets will be used from now on only as in (3.3). These should also not be confused with the brackets used in the introduction, there for mere simplicity. Furthermore, i is the imaginary unit. That symbol will not be used as an index, so that there should be no confusion here either.

which will take care of the finite self-energy terms. Furthermore, $\phi(\mathbf{r})$ will stand for $\Phi(f)$ if $f \to \delta(\mathbf{r})$, which is allowed since V is continuous by assumption. In that case,

$$:e^{i\phi(\mathbf{r})}: = e^{(1/2)\beta V(0)} e^{i\phi(\mathbf{r})} \tag{3.5}$$

In addition, define

$$\Upsilon_\Lambda[\phi] = \int_\Lambda :e^{i\phi(\mathbf{r})}: \kappa(d^3 r) \tag{3.6}$$

The argument ϕ and the subscript Λ will be dropped in most of the following, except when it seems appropriate to emphasize one or the other dependence. Clearly,

$$|\Upsilon|^2 = |\Upsilon^*|^2 = \Upsilon\Upsilon^* \tag{3.7}$$

where Υ^* means complex conjugate of Υ.

For simplicity, let the negative particles have odd integers as indices and the positive particles even ones. Thus $V_{k,j} = (-1)^{k+j} V$. If one now chooses $f \to \sum_{k=1}^{2N} (-1)^k \delta(\mathbf{r}_k)$, one obtains

$$\exp(-\beta U^{(N)}) = \left\langle \prod_{k=1}^{2N} :\exp[(-1)^{k+1} i\phi(\mathbf{r}_k)]: \right\rangle \tag{3.8}$$

Integration over all coordinates gives, after exchanging the function-space integrals with the configuration-space ones, a compact expression for (2.4b),

$$Z(N, \Lambda) = N!^{-2} \langle (\Upsilon_\Lambda \Upsilon_\Lambda^*)^N \rangle = N!^{-2} \langle |\Upsilon_\Lambda|^{2N} \rangle \tag{3.9}$$

4. BOUNDS FOR THE CLASSICAL CORRELATION FUNCTIONS

In this section the uniform upper bounds for the classical correlation functions will be proven. Recall that

$$\rho_{n_+,n_-}^{(N,\Lambda)} = c(N; n_+, n_-)\, \eta_{n_+,n_-}^{(N,\Lambda)} \tag{4.1}$$

with $\eta_{n_+,n_-}^{(N,\Lambda)} \in L^\infty(\Lambda^{n_+ + n_-})$ being a marginal probability density of (2.4). For further convenience, let us introduce some notation. For any $\mathcal{N} \subset \mathbb{N}$, let $|\mathcal{N}| = \mathrm{card}(\mathcal{N})$. Set $\mathscr{C} = \{1, 2,..., 2N\} \subset \mathbb{N}$, and let $\mathcal{N}_\pm \subset \mathscr{C}$ with $|\mathcal{N}_\pm| = n_\pm \leqslant N$ contain only integers of the index set of the positive, respectively negative, species. In terms of the representation introduced in

Section 2, the densities of the marginal canonical measures are then given by

$$\eta_{n_+,n_-}^{(N,\Lambda)} = \langle |\Upsilon_\Lambda|^{2N} \rangle^{-1} \langle \mathscr{F}_{n_+,n_-} \Upsilon_\Lambda^{N-n_+} \Upsilon_\Lambda^{*\,N-n_-} \rangle \tag{4.2a}$$

$$\mathscr{F}_{n_+,n_-}(\phi) = \prod_{j \in \mathscr{N}_+} :e^{i\phi(\mathbf{r}_j)}: \prod_{k \in \mathscr{N}_-} :e^{-i\phi(\mathbf{r}_k)}: \tag{4.2b}$$

Proposition 4.1. The configurational correlation functions $\rho_{n_+,n_-}^{(N,\Lambda)}$ of the classical canonical ensemble for charge-symmetric, neutral systems are bounded from above by

$$\rho_{n_+,n_-}^{(N,\Lambda)} \leqslant \rho_0^{2\bar{n}} e^{\bar{n}\beta V(0)}$$

where $\bar{n} \equiv A(n_+, n_-) = (1/2)(n_+ + n_-)$ is the arithmetic mean of n_+ and n_-, and $\rho_0 = \lambda_{\mathrm{dB}}^3 N/|\Lambda|$.

Remark. A canonical T-limit sequence is defined by an increasing mapping $N \mapsto \Lambda(N)$, with $N/|\Lambda|$ fixed, and $\Lambda \uparrow \mathbb{R}^3$. The bounds given by Proposition 4.1 are uniform along any such sequence.

To prove Proposition 4.1, the following lemma is useful.

Lemma 4.1. Let $(X, \mathscr{B}, d\pi)$ be a probability measure space, and $G: X \to \mathbb{R}^+$ in $L^p(X, d\pi)$ for all $p \in (0, P)$ (P might be ∞). Then for any two positive reals a, b, with $a + b \leqslant P$, the following inequality for expectations of powers of G holds:

$$E_\pi(G^{a+b}) \geqslant E_\pi(G^a)\, E_\pi(G^b)$$

The following is a very simple proof.

Proof of Lemma 4.1. Abbreviate $a + b = c$, and assume without loss of generality that $a \leqslant b$. For $\xi \in \mathbb{R}^+$, the mappings $\xi \mapsto \xi^{c/b}$ and $\xi \mapsto \xi^{b/a}$ are convex. Hence,

$$\begin{aligned} E_\pi(G^c) &= E_\pi(G^{b(c/b)}) \geqslant [E_\pi(G^b)]^{c/b} = E_\pi(G^b)[E_\pi(G^b)]^{a/b} \\ &= E_\pi(G^b)[E_\pi(G^{a(b/a)})]^{a/b} \geqslant E_\pi(G^b)\, E_\pi(G^a) \end{aligned}$$

by applying Jensen's inequality two times. ▮

Remark. There are other ways of proving this lemma. It may be considered as a special case of the more general inequality for simultaneously increasing functions, which was pointed out to me by J. Percus. A more indirect way is to prove it as a corollary of the generalized version of Theorem 197 of ref. 17 on convexity of the logarithm of power means, which was pointed out to me by B. Braams.

Proof of Proposition 4.1. The first step is to estimate $N!^2 Z\eta^{(N,\Lambda)}_{n_+,n_-}$ from above with the help of $|\langle f\rangle| \leqslant \langle |f|\rangle$, combined with (3.5) and (3.7). Using (4.2), this yields

$$\langle \mathscr{F}_{n_+,n_-} \Upsilon^{N-n_+} \Upsilon^{*\,N-n_-}\rangle \leqslant e^{\bar{n}\beta V(0)} \langle |\Upsilon|^{2N-2\bar{n}}\rangle$$

The second step is to use Lemma 4.1 for estimating

$$\langle |\Upsilon|^{2N}\rangle \geqslant \langle |\Upsilon|^{2N-2\bar{n}}\rangle \langle |\Upsilon|^{2\bar{n}}\rangle$$

The combination of steps one and two gives

$$\eta^{(N,\Lambda)}_{n_+,n_-} \leqslant e^{\bar{n}\beta V(0)} \langle |\Upsilon|^{2\bar{n}}\rangle^{-1}$$

Application of Jensen's inequality now yields

$$\langle |\Upsilon|^{2\bar{n}}\rangle \geqslant \langle |\Upsilon|\rangle^{2\bar{n}}$$

With

$$\langle |\Upsilon_\Lambda|\rangle \geqslant \langle \Upsilon_\Lambda\rangle = \int_\Lambda \kappa(d^3r) \int d\gamma(\phi)\, :e^{i\phi(\mathbf{r})}: = \lambda_{\mathrm{dB}}^{-3}\, |\Lambda|$$

(by the elementary properties of the Gaussian integrals as described in Section 3), and with

$$c(N; n_+, n_-) \leqslant N^{n_+ + n_-}$$

the proof of Proposition 4.1 is complete. ∎

For the strictly neutral grand canonical ensemble the corresponding estimate is done as follows. Let I_l be the modified Bessel function[18] of order l, with $l \in \mathbb{N} \cup \{0\}$. Using the representation in terms of Gaussian functional integrals for the marginal canonical probability densities, the $\sum_N$ for the partition function can immediately be carried out and reads

$$\Xi(\Lambda) = \langle I_0(2z\,|\Upsilon_\Lambda|)\rangle \tag{4.3}$$

which is readily verified by expanding the modified Bessel function I_0 into its Taylor series. The correlation function (2.6a) becomes

$$\varrho^{(\Lambda)}_{n_+,n_-} = \Xi(\Lambda)^{-1} \sum_{N=\bar{n}}^{\infty} z^{2N} \left\langle \mathscr{F}_{n_+,n_-} \frac{\Upsilon_\Lambda^{N-n_+}}{(N-n_+)!} \frac{\Upsilon_\Lambda^{*\,N-n_-}}{(N-n_-)!}\right\rangle \tag{4.4}$$

with $\mathscr{F}_{n_+,n_-}$ given in (4.2b). The sum in (4.4) can be carried out as well, to yield a more compact expression for (4.4); however, that is not as immediate as for the partition function (see the next section). For our present purposes (4.4) is already fully satisfactory to allow for a simple proof of the following result.

Proposition 4.2. The configurational correlation functions $\varrho^{(\Lambda)}_{n_+,n_-}$ of the strictly neutral classical grand canonical ensemble for charge-symmetric systems are uniformly bounded from above by

$$\varrho^{(\Lambda)}_{n_+,n_-} \leqslant z^{2\bar{n}} e^{\bar{n}\beta V(0)} = e^{\bar{n}\beta[V(0)+2\mu]}$$

with $\bar{n}$ as defined in Proposition 4.1.

For the proof we recall an integral representation of modified Bessel functions,[18]

$$I_{|k|}(x) = \frac{1}{\pi} \int_0^\pi e^{x \cos\theta} \cos(k\theta)\, d\theta \tag{4.5}$$

for $k \in \mathbb{Z}$.

Proof of Proposition 4.2. Since all individual terms in the series (4.4) are positive, an upper bound is obtained by replacing the integrands of the functional averages in (4.4) by their absolute values. Using (3.5) and furthermore (3.7), we find the estimate

$$\varrho^{(\Lambda)}_{n_+,n_-} \leqslant z^{2\bar{n}} e^{\bar{n}\beta V(0)} \frac{\langle I_{|q|}(2z\,|\Upsilon|)\rangle}{\langle I_0(2z\,|\Upsilon|)\rangle}$$

where $q = n_+ - n_-$ is the net charge number of the correlation function. Again, this is readily verified by expanding $I_{|q|}$ into its Taylor series.

The claim now follows from the pointwise inequality $I_0(x) - I_{|q|}(x) \geqslant 0$ for $x \in \mathbb{R}^+$, which in turn is easily shown by using the integral representation (4.5) for the cases $k = 0$ and $k = |q|$, and furthermore noting that $1 - \cos(\cdot) \geqslant 0$. ▮

The special case $n_+ = n_- = n$, so that $n = \check{n} = \bar{n}$, deserves additional attention. The correlation functions (2.6a) now obviously reduce to

$$\begin{aligned} \varrho^{(\Lambda)}_{n,n} &= z^{2n} \langle I_0(2z\,|\Upsilon|)\rangle^{-1} \left\langle \prod_{k=1}^{2n} :e^{(-1)^{k+1} i\phi(\mathbf{r}_k)}:\, I_0(2z\,|\Upsilon|)\right\rangle \\ &\equiv \left[\!\left[z^{2n} \prod_{k=1}^{2n} :e^{(-1)^{k+1} i\phi(\mathbf{r}_k)}: \right]\!\right] \end{aligned} \tag{4.6}$$

where for simplicity $\mathcal{N}_+ \cup \mathcal{N}_- = \{1, 2,..., 2n\}$ has been chosen. The expectation functional $[\![\cdot]\!]$ is defined by (4.6). It follows immediately from $|[\![f]\!]| \leqslant [\![|f|]\!]$ and (3.5) that

$$\varrho_{n,n}^{(\Lambda)} \leqslant z^{2n} e^{n\beta V(0)} \tag{4.7}$$

The bounds of the neutral correlation functions of the strictly neutral GCE are obtained like the general bounds in the mean-neutral unrestricted GCE.[2] Note that the expectation functional $[\![\cdot]\!]$ does not appear to have the structure of a thermal average; however, $[\![\cdot]\!]$ is a sine-Gordon measure in the sense of Kennedy.[19] Consequently, the correlation inequalities proved in ref. 19, Section 5, are valid for the expectation functional $[\![\cdot]\!]$ as well.

5. INCREASE OF THE GRAND CANONICAL CORRELATION FUNCTIONS

Proving the increase of the finite-volume correlation functions along the T-limit sequence turns out to be an elusive undertaking in the representation (4.1)–(4.2) and (4.3)–(4.4). However, (4.1)–(4.2) and (4.3)–(4.4) can be brought into a more convenient form. At least for the strictly neutral GCE, the proof of the increase of the correlation functions is then straightforward. For the CE, see the remarks in Appendix B.

First recall some simple algebra, namely

$$(e^{i\varphi} + e^{-i\varphi})^M = \sum_{k=0}^{M} \binom{M}{k} e^{i(M-2k)\varphi} \tag{5.1}$$

Upon integrating (5.1) over an interval of length 2π, all terms in the sum integrate to zero unless $M = 2N$, in which case the term with $k = N$ contributes the value $2\pi(2N)!/N!^2$. Along the same lines we find

$$N!^{-2}\,|\Upsilon|^{2N} = (2N)!^{-1}\,\frac{1}{2\pi}\int_{-\pi}^{\pi} d\varphi \langle (\Upsilon e^{i\varphi} + \Upsilon^* e^{-i\varphi})^{2N} \rangle \tag{5.2}$$

We define

$$\Theta_\Lambda[\phi; \varphi] \equiv 2 \int_\Lambda :\cos[\phi(\mathbf{r}) + \varphi]: \kappa(d^3r) = \Upsilon_\Lambda[\phi]\, e^{i\varphi} + \Upsilon_\Lambda^*[\phi]\, e^{-i\varphi} \tag{5.3}$$

The variables Λ, ϕ, and φ will occasionally be dropped when there is no risk of confusion. Introducing the abbreviation $(1/2\pi) \int_{-\pi}^{\pi} g(\varphi)\, d\varphi = \bar{g}$, we can write the neutral canonical partition function (3.9) as

$$Z(N, \Lambda) = (2N)!^{-1}\, \overline{\langle \Theta_\Lambda^{2N} \rangle} \tag{5.4}$$

The strictly neutral grand canonical partition function (2.6b) becomes

$$\Xi(\Lambda) = \overline{\langle \cosh(z\Theta_\Lambda)\rangle} \tag{5.5a}$$

By the above-mentioned fact that the φ averages of odd powers of Θ vanish, we have $\overline{\langle \sinh(z\Theta)\rangle} = 0$. Hence (5.5a) is identical to

$$\Xi(\Lambda) = \overline{\langle \exp(z\Theta_\Lambda)\rangle} \tag{5.5b}$$

which is easier to handle.

Following the same pattern, the correlation functions for the stictly neutral ensembles similarly can be brought into a more convenient form. Defining

$$\mathscr{D}_{n_+,n_-}(\phi;\varphi) \equiv \prod_{j\in\mathscr{N}_+} :e^{i(\phi(\mathbf{r}_j)+\varphi)}: \prod_{k\in\mathscr{N}_-} :e^{-i(\phi(\mathbf{r}_k)+\varphi)}: \tag{5.6}$$

(the arguments ϕ and φ will be dropped from $\mathscr{D}_{n_+,n_-}$ occasionally) we obtain from (4.1)–(4.2)

$$\rho^{(N,\Lambda)}_{n_+,n_-} = Z(N,\Lambda)^{-1}\,(2[N-\bar{n}])!^{-1}\,\overline{\langle \mathscr{D}_{n_+,n_-}\Theta_\Lambda^{2[N-\bar{n}]}\rangle} \tag{5.7}$$

and (4.4) becomes

$$\varrho^{(\Lambda)}_{n_+,n_-} = \Xi(\Lambda)^{-1}\overline{\langle z^{2\bar{n}}\mathscr{D}_{n_+,n_-}\exp(z\Theta_\Lambda)\rangle} \tag{5.8}$$

Upon expanding the exponential function in (5.8) into a Taylor series, formally one generates additional terms, as compared to (4.4). All additional terms average to zero, however, for the reasons mentioned at the beginning of this section.

We may now notice that the representation (5.8) has the form of a formal thermal average of a continuum plane rotator model. We can thus summarize: The strictly neutral GCE is obtained from the mean neutral one via the replacements $\phi(\mathbf{r}) \to \phi(\mathbf{r}) + \varphi$ and $\langle\cdot\rangle \to \overline{\langle\cdot\rangle}$.

At this point it is clear that one can prove the increase of the correlation functions of the strictly neutral GCE by the same techniques as used by Fröhlich and Park. We will give a slightly different proof below that does not need the interpolation argument of ref. 2. We note further that Proposition 4.2 now follows directly by the method of ref. 2, alternatively. However, the proof of Proposition 4.1 does not seem to become any simpler after the transformation into the new representation.

Proposition 5.1. The finite-volume correlation functions of the strictly neutral grand canonical ensemble are pointwise increasing along any increasing sequence $\Lambda \nearrow \mathbb{R}^3$, i.e., for any $\Lambda' \supset \Lambda$ (with $\Lambda \neq \Lambda'$) we find

$$\varrho^{(\Lambda')}_{n_+,n_-} > \varrho^{(\Lambda)}_{n_+,n_-}$$

Proof of Proposition 5.1. The proof is patterned after a theme of Ginibre.[7] We have

$$\begin{aligned}
&\Xi(\Lambda)\,\Xi(\Lambda')(\varrho^{(\Lambda')}_{n_+,n_-}-\varrho^{(\Lambda)}_{n_+,n_-})\\
&\quad=\overline{\langle z^{2\bar n}\mathscr{D}_{n_+,n_-}\{\exp(z\Theta_{\Lambda'})\,\Xi(\Lambda)-\exp(z\Theta_\Lambda)\,\Xi(\Lambda')\}\rangle}\\
&\quad=\mathrm{Ave}(z^{2\bar n}\mathscr{D}_{n_+,n_-}(\phi;\varphi)\{\exp(z\Theta_{\Lambda'}[\phi;\varphi])\exp(z\Theta_\Lambda[\tilde\phi;\tilde\varphi])\\
&\qquad-\exp(z\Theta_\Lambda[\phi;\varphi])\exp(z\Theta_{\Lambda'}[\tilde\phi;\tilde\varphi])\})
\end{aligned}$$

where

$$\mathrm{Ave}(\cdot)=\frac{1}{2\pi}\int_{-\pi}^{\pi}d\varphi\,\frac{1}{2\pi}\int_{-\pi}^{\pi}d\tilde\varphi\int d\gamma(\Phi)\int d\gamma(\tilde\Phi)(\cdot)$$

The term in curly braces can be rewritten as

$$\begin{aligned}
&\exp\{z(\Theta_\Lambda[\phi;\varphi]+\Theta_\Lambda[\tilde\phi;\tilde\varphi])\}\\
&\qquad\times[\exp(z\Theta_{\Lambda'\setminus\Lambda}[\phi;\varphi])-\exp(z\Theta_{\Lambda'\setminus\Lambda}[\tilde\phi;\tilde\varphi])]\\
&\quad=2\exp\{z(\Theta_\Lambda[\phi;\varphi]+\Theta_\Lambda[\tilde\phi;\tilde\varphi])\}\\
&\qquad\times\exp\{(z/2)(\Theta_{\Lambda'\setminus\Lambda}[\phi;\varphi]+\Theta_{\Lambda'\setminus\Lambda}[\tilde\phi;\tilde\varphi])\}\\
&\qquad\times\sinh\{(z/2)(\Theta_{\Lambda'\setminus\Lambda}[\phi;\varphi]-\Theta_{\Lambda'\setminus\Lambda}[\tilde\phi;\tilde\varphi])\}
\end{aligned}$$

The terms in the curly braces will now be transformed using an orthogonal transformation of the field variables ϕ, $\tilde\phi$, an orthogonal plus stretching transformation for the variables φ and $\tilde\varphi$, as well as trigonometric identities. Define

$$\frac{\phi+\tilde\phi}{\sqrt2}=\psi\qquad\frac{\varphi+\tilde\varphi}{2}=\alpha$$

$$\frac{-\phi+\tilde\phi}{\sqrt2}=\tilde\psi\qquad\frac{-\varphi+\tilde\varphi}{2}=\tilde\alpha$$

Under these transformations, the measures and domains of integration transform as[2]

$$\int d\gamma(\Phi)\int d\gamma(\tilde\Phi)(\cdot)=\int d\gamma(\Psi)\int d\gamma(\tilde\Psi)(\cdot)$$

where $\Psi(\delta[\mathbf r])=\psi(\mathbf r)$, and, for periodic integrands,[7]

$$\frac{1}{2\pi}\int_{-\pi}^{\pi}d\varphi\,\frac{1}{2\pi}\int_{-\pi}^{\pi}d\tilde\varphi(\cdot)=\frac{1}{2\pi}\int_{-\pi}^{\pi}d\alpha\,\frac{1}{2\pi}\int_{-\pi}^{\pi}d\tilde\alpha(\cdot)$$

The arguments of the exponential functions and of the hyperbolic sine are rewritten by using the identities

$$\cos(\phi+\varphi)+\cos(\tilde{\phi}+\tilde{\varphi})=2\cos(\sqrt{\tfrac{1}{2}}\,\psi+\alpha)\cos(\sqrt{\tfrac{1}{2}}\,\tilde{\psi}+\tilde{\alpha})$$
$$\cos(\phi+\varphi)-\cos(\tilde{\phi}+\tilde{\varphi})=2\sin(\sqrt{\tfrac{1}{2}}\,\psi+\alpha)\sin(\sqrt{\tfrac{1}{2}}\,\tilde{\psi}+\tilde{\alpha})$$

We observe further that only the real part of $\mathscr{D}_{n_+,n_-}$ contributes to (5.7). We have

$$\mathrm{Re}[\mathscr{D}_{n_+,n_-}(\phi;\varphi)] =: \cos\left\{\sum_{k\in\mathscr{N}_+\cup\mathscr{N}_-}\sigma_k[\phi(\mathbf{r}_k)+\varphi]\right\}:$$

where $\sigma_k=\pm 1$ for $k\in\mathscr{N}_\pm$. This can be transformed using the identity

$$\cos(\phi+\varphi)=\cos(\sqrt{\tfrac{1}{2}}\,\psi+\alpha)\cos(\sqrt{\tfrac{1}{2}}\,\tilde{\psi}+\tilde{\alpha})$$
$$+\sin(\sqrt{\tfrac{1}{2}}\,\psi+\alpha)\sin(\sqrt{\tfrac{1}{2}}\,\tilde{\psi}+\tilde{\alpha})$$

which applies equally well if $\phi+\varphi$ is replaced by a sum of the form $\sum_k\sigma_k[\phi+\varphi]$.

The rest is the standard argument of Ginibre. We expand the exponential functions and the hyperbolic sine into their Taylor series, which have only positive Taylor coefficients. In the resulting multiple sum we exchange the field integrations and the phase space integrations in each term. The field integrations factorize into products of two identical real integrals, which are thus positive. Hence, the total sum is positive. ▮

Theorem 5.1. The limit

$$\lim_{\Lambda\nearrow\mathbb{R}^3}\varrho^{(\Lambda)}_{n_+,n_-}=\varrho_{n_+,n_-}$$

for the correlation functions of the strictly neutral charge-symmetric grand canonical ensemble exists along any increasing sequence of domains Λ, and for all real nonnegative β and z. The limit is independent of the sequence of domains and is given by the variational formula

$$\varrho_{n_+,n_-}=\max_{\Lambda}\varrho^{(\Lambda)}_{n_+,n_-}$$

which holds pointwise.

Proof of Theorem 5.1. As a corollary of Propositions 4.2 and 5.1, we have existence of the limit along any increasing sequence of domains, and also pointwise convergence to the maximum. Uniqueness can be seen as follows. Let $s\in\mathbb{R}^+$ and let $s\mapsto\Lambda'(s)$ and $s\mapsto\Lambda''(s)$ be two nonidentical increasing sequences of domains that converge to $\mathbb{R}^3$. We can then construct a third increasing sequence $s \mapsto \Lambda(s)$ which "alternates" between these two, such that we can extract increasing subsequences $n \mapsto \Lambda_a(s[n])$ and $n \mapsto \Lambda_b(s[n])$ of $\Lambda(s)$, with $n \in \mathbb{N}$, and $\Lambda_a(s[n])$ contains only elements which are in $\Lambda'(s)$, while $\Lambda_b(s[n])$ contains only elements which are in $\Lambda''(s)$. Since the limit of the correlation functions along $\Lambda(s)$ and along any of its subsequences must be the same, we see that the limit is the same along $\Lambda'(s)$ and $\Lambda''(s)$, and thus for all increasing sequences. ∎

We conclude this section with a few remarks regarding further properties of the correlation functions. Since V is translationally and rotationally invariant [see (2.3)], the correlation functions have these properties in the thermodynamic limit.[20] Moreover, since the limit exists for all real nonnegative β and z, we can rule out the existence of a first-order phase transition. Recall that at a first-order phase transition the correlation functions have a convex continuum of limit points instead of a limit. The extreme points of that continuum are the pure phases; all other points represent mixed phases.

APPENDIX A. QUANTUM SYSTEMS OF DISTINGUISHABLE PARTICLES

In the following we summarize how essentially the same formalism goes through in the case of quantum systems with Boltzmann statistics. Only the basic ingredients are presented. For further details on the representation see the articles by Siegert[6] and Fröhlich and Park[2] and also the books by Glimm and Jaffe[13] and Simon.[16]

The Hilbert space for a finite neutral system is $\mathscr{H}_\Lambda^{(N)} = \bigotimes_{j=1}^{2N} L^2(\Lambda, d^3r_j)$. The Hamiltonian $H^{(N)}$ is as in (2.1), with $m_j = m$ for all j, and with $|\mathbf{p}_j|^2 = -\hbar^2\nabla_j^2$ for 0-Dirichlet data at the boundary of Λ_j. Then $\exp(-\beta H^{(N)})$ is trace class for $\beta > 0$. Let $P_{\Lambda,\beta}^{(N)}(\mathbf{r}_1,..., \mathbf{r}_{2N}; \mathbf{r}'_1,..., \mathbf{r}'_{2N})$ be the resolvent kernel of $\exp(-\beta H^{(N)})$ on $\Lambda^{2N} \times \Lambda^{2N}$, and define

$$\chi_\Lambda^{(N)}(\mathbf{r}_1,..., \mathbf{r}_{2N}) \equiv \int_{\Lambda^{2N}} P_\Lambda^{(N)} \bigotimes_{k=1}^{2N} \delta(\mathbf{r}_k - \mathbf{r}'_k) \prod_{j=1}^{2N} d^3r'_j \tag{A.1}$$

Then $\hat{Q}(N, \Lambda)^{-1} \chi_\Lambda^{(N)}$ is the quantum analog of (2.4a), with $\hat{Q}(N, \Lambda) = \int_{\Lambda^{2N}} \chi_\Lambda^{(N)} d^{6N}r$.

The Siegert representation of (A.1) that is analogous to (3.8) is obtained from the Feynman–Kac formula[13,16] and the subsequent application of the Gaussian functional representation. Let $\dot{P}_{\Lambda,\beta}(\mathbf{r}, \mathbf{r}'; d\omega)$ be the path space measure on $\times_{\tau \in [0,\beta]} \dot{\mathbb{R}}_\tau^3$ for the Wiener process with transition function $\exp(\tau\hbar^2\nabla^2/2m)$ conditioned by $\omega(\tau = 0) = \mathbf{r}$ and $\omega(\tau = \beta) = \mathbf{r}'$, with $\mathbf{r}$ and $\mathbf{r}'$ in Λ. Here, $\dot{\mathbb{R}}^3$ is the one-point compactification of $\mathbb{R}^3$.[16] Let $U^{(N)}(\tau)$ be given by (2.3) with $\mathbf{r}_j$ replaced by $\omega_j(\tau)$ for all $j = 1, 2, \ldots, 2N$. The Feynman–Kac formula represents the resolvent kernel $P^{(N)}_{\Lambda,\beta}$ as

$$\begin{aligned} &P^{(N)}_{\Lambda,\beta}(\mathbf{r}_1, \ldots, \mathbf{r}_{2N}; \mathbf{r}'_1, \ldots, \mathbf{r}'_{2N}) \\ &\quad = \int_{\Omega^{2N}} \prod_{j=1}^{2N} \dot{P}_{\Lambda,\beta}(\mathbf{r}_j, \mathbf{r}'_j; d\omega_j) \exp\left[-\int_0^\beta d\tau\, U^{(N)}(\tau) \right] \end{aligned} \tag{A.2}$$

from which the corresponding representation for χ follows.

The Hilbert spaces $\mathscr{H}_{\pm\infty}$ are now constructed from $L^2(\mathbb{R}^3 \times [0, \beta], d^3r\, d\tau)$ instead of $L^2(\mathbb{R}^3, d^3r)$ as in the classical case. The kernel of the corresponding bilinear form is $\mathscr{V} = V(\mathbf{r} - \mathbf{r}')\, \delta(\tau - \tau')$. This gives[2,6]

$$\exp\left[-\int_0^\beta d\tau\, U^{(N)}(\tau) \right] = \left\langle \prod_{j=1}^{2N} :\!\exp\left\{ i(-1)^{j+1} \int_0^\beta d\tau\, \phi(\omega_j[\tau]; \tau) \right\}\!: \right\rangle_q \tag{A.3}$$

where $\langle \cdot \rangle_q$ denotes the average with the Gaussian measure with covariance $\mathscr{V}$ and mean 0. The Wick ordering is the same as in the classical case. (Notice that in the present paper the interaction V depends only on the relative distance between particles. For more general V see Fröhlich and Park.[2])

It is now obvious that in the quantum case the same structure is obtained for the expressions of the correlation functions as in the classical case if one makes the replacements

$$:\!\exp[\pm i\phi(\mathbf{r})]\!: \rightarrow \int_\Omega \dot{P}_{\Lambda,\beta}(\mathbf{r}, \mathbf{r}; d\omega) :\!\exp\left\{ \pm i \int_0^\beta d\tau\, \phi(\omega[\tau]; \tau) \right\}\!: \tag{A.4a}$$

$$\langle \cdot \rangle \rightarrow \langle \cdot \rangle_q \tag{A.4b}$$

In particular, with the abbreviation

$$\hat{\Upsilon}_\Lambda(\phi) \equiv \int_\Lambda d^3r \int_\Omega \dot{P}_{\Lambda,\beta}(\mathbf{r}, \mathbf{r}; d\omega) :\!\exp\left\{ i \int_0^\beta d\tau\, \phi(\omega[\tau]; \tau) \right\}\!: \tag{A.5}$$

the canonical partition function $\hat{Z}$ becomes

$$\hat{Z}(N, \Lambda) = N!^{-2} \operatorname{Tr} e^{-\beta H^{(N)}} = N!^{-2} \langle |\hat{\Upsilon}_\Lambda|^{2N} \rangle_q \tag{A.6}$$

The restricted grand canonical partition function is

$$\hat{\Xi}(\Lambda) = \sum_{N=0}^{\infty} z^{2N} \hat{Z}(N, \Lambda) = \langle I_0(2z\, |\hat{\Upsilon}_\Lambda|) \rangle_q \tag{A.7}$$

For the sake of brevity, the (now obvious) expressions for the Siegert representation of the particle density correlation functions are omitted. The φ-averaged Siegert representation will be given below.

Proposition A.1. The quantum mechanical particle density correlation functions $\hat{\rho}^{(\Lambda,N)}_{n_+,n_-}$ and $\varrho^{(\Lambda)}_{n_+,n_-}$, defined by the replacements (A.4a)–(A.4b) in (4.1)–(4.2) and (4.3)–(4.4), respectively, are bounded by

$$\hat{\rho}^{(\Lambda,N)}_{n_+,n_-} \leqslant \rho_0^{2\bar{n}} e^{\bar{n}\beta V}$$

and

$$\hat{\varrho}^{(\Lambda)}_{n_+,n_-} \leqslant z^{2\bar{n}} e^{\bar{n}\beta V}$$

as in the classical case.

Proof of Proposition A.2. One has to copy the steps given in the proofs of Propositions 4.1 and 4.2, except for one additional estimate,

$$\int_\Omega \dot{P}_{\Lambda,\beta}(\mathbf{r}, \mathbf{r}; d\omega) \leqslant \int_\Omega \dot{P}_{\mathbb{R}^3,\beta}(\mathbf{r}, \mathbf{r}; d\omega) = (2\pi m/\beta h^2)^{3/2}$$

which is standard (see, for instance, ref. 2). This estimate has to be amended in the first step in either of the proofs of the classical propositions. ∎

Next we define

$$\hat{\Theta}_\Lambda[\phi; \varphi] = \hat{\Upsilon}_\Lambda[\phi]\, e^{i\varphi} + \hat{\Upsilon}^*_\Lambda[\phi]\, e^{-i\varphi} \tag{A.8}$$

which is the quantum analog of (5.3), and

$$\begin{aligned} \hat{\mathscr{D}}_{n_+,n_-}(\phi; \varphi) = \prod_{k \in \mathscr{N}_+ \cup \mathscr{N}_-} & \int_\Omega \dot{P}_{\Lambda,\beta}(\mathbf{r}_k, \mathbf{r}_k; d\omega_k) \\ & \times :\exp\left\{ i\sigma_k \left[\int_0^\beta \phi(\omega_k[\tau]; \tau)\, d\tau + \varphi \right] \right\}: \end{aligned} \tag{A.9}$$

We concentrate now only on the grand canonical correlation functions. Analogous to the classical case, we obtain

$$\hat{\Xi}(\Lambda) = \overline{\langle \exp(z\hat{\Theta}_\Lambda) \rangle_q} \tag{A.10}$$

and

$$\hat{\varrho}^{(\Lambda)}_{n_+,n_-} = \hat{\Xi}(\Lambda)^{-1}\, \overline{\langle z^{2\bar{n}} \hat{\mathscr{D}}_{n_+,n_-} \exp(z\hat{\Theta}_\Lambda) \rangle_q} \tag{A.11}$$

We can now state the following result.

Proposition A.2. The quantum mechanical finite-volume particle density correlation functions of the strictly neutral grand canonical ensemble are pointwise increasing along any increasing sequence $\Lambda \nearrow \mathbb{R}^3$, i.e.,

$$\hat{\varrho}^{(\Lambda')}_{n_+,n_-} > \hat{\varrho}^{(\Lambda)}_{n_+,n_-}$$

for any $\Lambda' \supset \Lambda$.

Theorem A.1. The limit

$$\lim_{\Lambda \nearrow \mathbb{R}^3} \hat{\varrho}^{(\Lambda)}_{n_+,n_-} = \hat{\varrho}_{n_+,n_-}$$

of the quantum mechanical density correlation functions of the strictly neutral charge-symmetric grand canonical ensemble exists along any increasing sequence of domains Λ, and for all real nonnegative β and z. The limit is independent of the sequence of domains and given by

$$\hat{\varrho}_{n_+,n_-} = \max_{\Lambda} \hat{\varrho}^{(\Lambda)}_{n_+,n_-}$$

which holds pointwise.

The proofs are essentially identical to those given in Section 5.

APPENDIX B. SUBADDITIVITY OF THERMODYNAMIC POTENTIALS

The closely related problem of the existence of the thermodynamic functions is now briefly commented upon. For the sake of brevity, only the classical systems are treated.

Fröhlich and Park[2] used the existence of the thermodynamic limit of the correlation functions for arbitrary increasing sequences of domains Λ to conclude likewise the existence of the thermodynamic limit of the grand canonical pressure, and of other related thermodynamic functions along arbitrary increasing sequences of domains. It is of interest to prove the existence of the thermodynamic limit of the thermodynamic functions without the recourse to the correlation functions.

If one is satisfied with proving this for sequences of standard cubes[20] (or more generally for van Hove sequences), a nice argument due to Griffiths[9] is sufficient to prove subadditivity of the canonical free energy, respectively superadditivity of the grand canonical potential. The argument applies to strictly as well as mean neutral systems.

The sub(super)additivity can be recovered in the Siegert representation along the lines of Section 5. For the mean neutral GCE this was done in ref. 2. For the strictly neutral GCE the proof is essentially identical to the one given in ref. 2, with only the slight modifications already needed in Section 5. For the neutral CE the proof is an interesting variant and will be given below.

It should be noted that Griffiths' proof of subadditivity is much simpler than the one based on the Siegert representation. The idea of giving the proof of subadditivity using the Siegert representation is more of conceptual interest, namely to have a base from which one may start in order to prove a stronger result: strong subadditivity of the canonical free energy (if this holds at all). For both grand canonical ensembles the corresponding result is strong superadditivity of the finite-volume grand potential. This can indeed be proved (see below) along the lines of Section 5, but it seems hard to prove it by an argument as simple as Griffiths'. The strong superadditivity allows one to prove the thermodynamic limit for more general increasing sequences of domains without invoking the correlation functions.

Proposition B.1 (Griffiths). The finite-volume canonical free energy $F(N, \Lambda) = -\beta^{-1} \log Z(N, \Lambda)$ of charge-symmetric systems is subadditive in the sense

$$F(N' + N, \Lambda' \cup \Lambda) \leqslant F(N', \Lambda') + F(N, \Lambda)$$

for $\Lambda' \cap \Lambda = \varnothing$, and N', N in $\mathbb{N}$.

Proof of Proposition B.1. Obviously it is sufficient to prove

$$Z(N' + N, \Lambda' \cup \Lambda) \geqslant Z(N', \Lambda')\, Z(N, \Lambda)$$

Using (5.4) and (5.3), we see that

$$\begin{aligned} Z(N' + N, \Lambda' \cup \Lambda) &= (2[N' + N])!^{-1}\, \overline{\langle \Theta_{\Lambda' \cup \Lambda}^{2(N'+N)} \rangle} \\ &= \sum_{k=0}^{N'+N} (2[N' + N - k])!^{-1}\, (2k)!^{-1}\, \overline{\langle \Theta_{\Lambda'}^{2(N'+N-k)} \Theta_{\Lambda}^{2k} \rangle} \end{aligned}$$

Odd powers do not occur, according to the remark following (5.1). It is easily seen (for instance, by exchanging the field averages and the space integrations) that each term in the sum is positive (unless either Λ' or Λ is the empty set, which is uninteresting). As such, we find a lower estimate for $Z(N' + N, \Lambda' \cup \Lambda)$ by keeping only the term with $k = N$ in the above sum.

In order not to overload the ensuing expressions, let us define the abbreviations

$$(2[N'+N])!^{-1}\,\Theta^{2(N'+N)}_{\Lambda'\cup\Lambda}=\mathscr{I}$$
$$(2N')!^{-1}\,\Theta^{2N'}_{\Lambda'}=\mathscr{J}$$
$$(2N)!^{-1}\,\Theta^{2N}_{\Lambda}=\mathscr{K}$$

Furthermore

$$\Theta_\Lambda[\phi;\varphi]+\Theta_\Lambda[\tilde{\phi};\tilde{\varphi}]=2X$$
$$\Theta_\Lambda[\phi;\varphi]-\Theta_\Lambda[\tilde{\phi};\tilde{\varphi}]=2Y$$

and finally

$$M!^{-1}X^M=\mathscr{L}(M),\qquad M!^{-1}Y^M=\mathscr{M}(M)$$

We then obtain the estimate

$$\begin{aligned}
&Z(N'+N,\Lambda'\cup\Lambda)-Z(N',\Lambda')\,Z(N,\Lambda)\\
&\quad=\langle\bar{\mathscr{I}}\rangle-\langle\bar{\mathscr{J}}\rangle\langle\bar{\mathscr{K}}\rangle\\
&\quad=\mathrm{Ave}(\mathscr{I}[\phi;\varphi]-\mathscr{J}[\phi;\varphi]\,\mathscr{K}[\tilde{\phi};\tilde{\varphi}])\\
&\quad\geqslant\mathrm{Ave}(\mathscr{J}[\phi;\varphi][\mathscr{K}[\phi;\varphi]-\mathscr{K}[\tilde{\phi};\tilde{\varphi}]])\\
&\quad=2\sum_{m=1}^{N}\mathrm{Ave}(\mathscr{J}[\phi;\varphi]\,\mathscr{L}(2[N-m]+1)\,\mathscr{M}(2m-1))\equiv A
\end{aligned}$$

where Ave has been defined in Section 5. Note that the last step is just

$$(X+Y)^{2N}-(X-Y)^{2N}=2\sum_{m=1}^{N}\binom{2N}{2m-1}X^{2[N-m]+1}Y^{2m-1}$$

Now notice that we can rewrite $\Theta[\phi;\varphi]$, X, and Y by using again the orthogonal (plus stretching) transformations for the ϕ and φ variables and the trigonometric identities, as in the proof of Proposition 5.1. Indeed, all ensuing steps are now the standard steps as described in ref. 7, and as already remarked in the proof of Proposition 5.1, yielding $A\geqslant 0$. ∎

Proposition B.2. The finite-volume grand potential $p_\Lambda|\Lambda|=\beta^{-1}\log\Xi(\Lambda)$ of the (mean or strictly) neutral charge-symmetric systems is strongly superadditive, i.e.,

$$\log\Xi(\Lambda'\cup\Lambda)+\log\Xi(\Lambda'\cap\Lambda)\geqslant\log\Xi(\Lambda')+\log\Xi(\Lambda)$$

Proof of Proposition B.2. We give only the initial steps up to the point from which the proof proceeds exactly as the one of Proposition 5.1. Also, only the strictly neutral version is formulated; the mean neutral one is basically the same by the replacements mentioned in Section 5.

We define

$$z\Theta_{\Lambda'\setminus(\Lambda'\cap\Lambda)}=\mathscr{P}$$
$$z\Theta_{\Lambda'\cap\Lambda}=\mathscr{Q}$$
$$z\Theta_{\Lambda\setminus(\Lambda'\cap\Lambda)}=\mathscr{R}$$

and rewrite

$$\begin{aligned}
&\Xi(\Lambda'\cup\Lambda)\,\Xi(\Lambda'\cap\Lambda)-\Xi(\Lambda')\,\Xi(\Lambda)\\
&\quad=\mathrm{Ave}\{\exp(z\Theta_{\Lambda'\cup\Lambda}[\phi;\varphi])\exp(z\Theta_{\Lambda'\cap\Lambda}[\tilde{\phi};\tilde{\varphi}])\\
&\qquad-\exp(z\Theta_{\Lambda'}[\phi;\varphi])\exp(z\Theta_{\Lambda}[\tilde{\phi};\tilde{\varphi}])\}\\
&\quad=\mathrm{Ave}\{\exp(\mathscr{P}[\phi;\varphi])\exp(\mathscr{Q}[\phi;\varphi]+\mathscr{Q}[\tilde{\phi};\tilde{\varphi}])\\
&\qquad\times\left[\exp(\mathscr{R}[\phi;\varphi])-\exp(\mathscr{R}[\tilde{\phi};\tilde{\varphi}])\right]\}\equiv B
\end{aligned}$$

The expression in the large square brackets is identical to

$$2\exp\{\tfrac{1}{2}(\mathscr{R}[\phi;\varphi]+\mathscr{R}[\tilde{\phi};\tilde{\varphi}])\}\sinh\{\tfrac{1}{2}(\mathscr{R}[\phi;\varphi]-\mathscr{R}[\tilde{\phi};\tilde{\varphi}])\}$$

We have arrived basically at the same structure as encountered in the proof of Proposition 5.1. The remaining steps that show $B\geqslant 0$ are now clear, and are omitted. ∎

We now see that, for the grand canonical potential, strong superadditivity and superadditivity require essentially the same kind of proof. (Superadditivity is obtained by setting $\Lambda'\cap\Lambda=\varnothing$.) The term representing the integration over $\Lambda'\cap\Lambda$ does not introduce new structures.

For the canonical ensemble, on the other hand, it seems that the proof of subadditivity does not have an immediate generalization to obtain strong subadditivity. The estimates needed to show increase of the correlation functions and the estimates needed to show strong subadditivity are almost the same. Hence, it seems promising to invest some further effort into this problem. Progress may, however, come slowly.

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