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# The one dimensional plasma revisited: the influence of periodic boundary conditions

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‘Dedicated to Philippe Choquard on his 60th anniversary’

**Abstract.** We study the one dimensional alternating charge ordered two component plasma with one dimensional coulombic interactions. We use free boundary conditions where the system is well known to remain bound into molecules at all couplings and we use periodic boundary conditions, where the system has a transition from a low coupling unbound phase to a high coupling bound phase. We discuss the correlation functions and pressure in both systems and the distribution of zeros of the grand canonical partition functions considered as functions of fugacity. The effect of the periodic boundary conditions is to include an extra long ranged weak potential in the Hamiltonian from the free boundary condition system. This extra potential resembles a Kac potential. We construct an exact Lebowitz–Penrose theory to show how this extra Kac potential gives rise to the phase transition in the periodic system.

## I. Introduction

While the existence of the thermodynamic limit for short ranged potentials has been well understood for some twenty-five years [1, 2], the situation is not so clear with long ranged potentials like Coulombic ones. The existence of the thermodynamic limit for such systems was established by the work on stability of Dyson and Lenard [3] and the rest of the proof was developed by Lieb and Lebowitz [4]. These results were obtained for systems in free boundary conditions. A related piece of work was that of Fisher and Lebowitz [5] who showed that for systems which interacted with a pair potential  $\phi(|\mathbf{r}|)$  which decayed at large  $|\mathbf{r}|$  faster than  $|\mathbf{r}|^{-3-\epsilon}$ , the thermodynamic limit for the free energy in periodic boundary conditions exists and is equal to that in free boundary conditions. The ubiquitous use of periodic boundary conditions in solid state physics and in computer simulation of dense matter make this question a very important one.

There is no proof in the literature about the existence of the thermodynamic limit in periodic boundary conditions for Coulombic systems in general. There is a result by Penrose and Smith [6] that the free energy density exists for a system

in which the particles interact via a Coulombic potential derived from a Poisson equation with a Neumann boundary condition at the edge of the container of the particles. For dipolar systems, some incomplete results of Smith and Perram [7] indicate that the shift in potential caused by changing the electrostatic boundary conditions from free boundary conditions to periodic boundary conditions may act in the manner of a Kac potential and thus be capable of introducing new phase transitions or moving other phase transition points.

This paper considers the one dimensional charge alternating two component plasma. In Section II we introduce the Hamiltonian in free and in periodic boundary conditions, results which are reasonably standard. In Section III we study the canonical free energy of the free boundary condition system and the grand canonical pressure and correlation functions. To do this we use a simple variant of the method of Takahashi [8] for one dimensional systems. We also introduce a canonical (fixed particle number) fixed polarisation ensemble and calculate the free energy density in that ensemble. In Section IV we repeat the exercise for the case of periodic boundary conditions, our method seeming rather simpler than those of Schotte and Truong [9] and Kardar [10] for the same system. In Section V we discuss sum rules for the correlation functions that we have found, thus identifying the dielectric nature of the phases involved in the systems. We then move to discuss the zeros of the grand canonical partition function in free and periodic boundary conditions in Section VI. We find significant differences between the two different boundary conditions that we use. In Section VII we discuss how to derive the periodic system results from the free boundary condition results by a Lebowitz–Penrose argument [11], showing for this system at least, that the difference in potential between the free boundary condition system and the periodic boundary condition system acts as a Kac potential. We discuss the more general application of such an idea in the concluding Section VIII.

## II. The Hamiltonian

We consider a system of  $2N$  particles on a line of length  $L$  with particle  $j$  having a charge  $q_j = Qe_j = Q(-1)^j$ ,  $1 \leq j \leq 2N$ . Here  $Q$  is some elementary charge having the dimension of ordinary one dimensional charges. We shall use  $\gamma = Q^2/kT$  throughout this paper as a useful parameter. The dimensionless coupling parameter that we shall introduce in our results will be  $\Gamma = Q^2/2\rho kT$ , where  $\rho$  is the density of particles (of both signs,  $\rho = 2N/L$ ) in the system,  $k$  is Boltzmann's constant and  $T$  is the absolute temperature. The energy of the system is

$$\mathcal{H} = \frac{1}{2}Q^2 \sum_{j=1}^{2N} e_j \Psi(x_j) \quad (2.1)$$

where  $\Psi$  is the solution of

$$\nabla^2 \Psi(x) = -2 \sum_{k=1}^{2N} e_k \delta(x - x_k), \quad (2.2)$$

the one dimensional Poisson equation for the system with particles at  $x_j \in \{-L/2, L/2\}$ ,  $1 \leq j \leq 2N$ .

In free boundary conditions this Hamiltonian becomes

$$\mathcal{H}_F = -\frac{1}{2}Q^2 \sum_{j=1}^{2N} \sum_{k=1}^{2N} e_j e_k |x_j - x_k|. \quad (2.3)$$

In periodic boundary conditions, the problem may be solved by expanding the solution  $\Psi$  in a Fourier series, identifying the coefficients and then consulting a reliable [11] list of Fourier series for the sum. Using the overall charge neutrality of the system, we can reduce this periodic Hamiltonian to

$$\mathcal{H}_{PBC} = -\frac{1}{2}Q^2 \sum_{j=1}^{2N} \sum_{k=1}^{2N} e_j e_k |x_j - x_k| - Q^2 M^2 / L \quad (2.4)$$

where

$$M = \sum_{j=1}^{2N} e_j x_j = \sum_{k=1}^N x_{2k} - x_{2k-1} \quad (2.5)$$

is (barring a factor  $Q$ ), the dipole moment of the configuration  $\{x_1, x_2, \dots, x_{2N}\}$  concerned.

In this work we consider the particles to be charge ordered. That is, we insist that the coordinates of the particles satisfy the inequalities

$$\begin{aligned} 0 &\leq x_1 \leq x_2 \\ x_{j-1} &\leq x_j \leq x_{j+1}, \quad 2 \leq j \leq 2N-1 \\ x_{2N-1} &\leq x_{2N} \leq L. \end{aligned} \quad (2.6)$$

It is a relatively easy but somewhat lengthy exercise to show that with these constraints,

$$\beta \mathcal{H}_F = \gamma M \quad (2.7)$$

and so we may see that with these constraints we also have

$$\beta \mathcal{H}_{PBC} = -\frac{\gamma}{L} \left( M - \frac{L}{2} \right)^2 + \gamma L / 4. \quad (2.8)$$

The canonical partition functions are both of the form

$$\mathbb{Z}_{2N}(L, T, X) = \int_0^L dx_{2N} \int_0^{x_{2N}} dx_{2N-1} \int_0^{x_{2N-1}} dx_{2N-2} \cdots \int_0^{x_2} dx_1 \exp(-\beta \mathcal{H}_X) \quad (2.9)$$

where the  $X$  refers to the electrostatic boundary conditions being used. In free boundary conditions it will be convenient to introduce a constant polarization density ensemble, where we restrict the configurations to that surface in phase space on which

$$M = Nm_0, \quad \text{or} \quad M = L\sigma, \quad (2.10)$$

so that  $\sigma$  is the polarisation density of the system.

The reasons for introducing this ensemble will only become apparent in the last section. In this ensemble the partition function is

$$\mathbb{Z}_{2N}(L, T, F, \sigma, \gamma) = \int_0^L dx_{2N} \int_0^{x_{2N}} dx_{2N-1} \int_0^{x_{2N-1}} dx_{2N-2} \cdots \int_0^{x_2} dx_1 \\ \times \delta(M - L\sigma) \exp(-\beta \mathcal{H}_F). \quad (2.11)$$

For these systems we also introduce the grand canonical partition functions

$$\Xi(\zeta, L, T) = \sum_{N=0}^{\infty} \zeta^{2N} \mathbb{Z}_{2N}(L, T) \quad (2.12)$$

so that the pressure and density are given by

$$p(\zeta)/kT = \chi(\zeta) = \lim_{L \rightarrow \infty} \frac{1}{L} \log \{\Xi(\zeta, L, T)\} \quad (2.13)$$

and

$$\rho(\zeta) = \zeta \frac{\partial \chi}{\partial \zeta}(\zeta). \quad (2.14)$$

Notice, because of the way we have defined  $\zeta$  that  $\rho$  is the number density of particles in the system. The number density of positive particles in the system is  $\rho/2$ , as is the number density of negative particles. The grand canonical distribution functions for each system have their usual definitions based on this grand canonical partition function.

### III. The free boundary condition systems

In free boundary conditions, the canonical partition function may be written

$$\mathbb{Z}_{2N}(L, T, X) = F(L, N, \gamma) \quad (3.1)$$

where

$$F(L, N, \gamma) = \int_0^L dx_{2N} \int_0^{x_{2N}} dx_{2N-1} \int_0^{x_{2N-1}} dx_{2N-2} \cdots \int_0^{x_2} dx_1 \\ \times \exp\left(-\sum_{j=1}^{2N} \gamma(-1)^j x_j\right). \quad (3.2)$$

We may evaluate  $F$  by taking a Laplace transform in  $L$  to obtain

$$\hat{F}(s, N, \gamma) = \frac{1}{s} \{s(s + \gamma)\}^{-N} \quad (3.3)$$

and we may then obtain the free energy per particle in the thermodynamic limit by inverting this Laplace transform using a steepest descent method. The free energy per particle in the thermodynamic limit is then

$$\frac{a(\rho, T)}{kT} - \log(\rho/\sqrt{2}) = -\frac{1}{2}\{1 - 2\Gamma + \{1 + 4\Gamma^2\}^{1/2}\} + \frac{1}{2} \log \{1 + \sqrt{1 + 4\Gamma^2}\} \quad (3.4)$$

The grand canonical partition function is easily evaluated as

$$\begin{aligned} \Xi(\zeta, L, T) &= \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} \frac{s(s+\gamma)e^{sL}}{s\{s^2 + \gamma s - \zeta^2\}} ds \\ &= \frac{\zeta^2 e^{-L\gamma/2}}{2\sqrt{[\zeta^2 + \gamma^2/4]}} \left\{ \frac{\exp\{L\sqrt{[\zeta^2 + \gamma^2/4]}\}}{\sqrt{[\zeta^2 + \gamma^2/4]} - \frac{\gamma}{2}} + \frac{\exp\{-L\sqrt{[\zeta^2 + \gamma^2/4]}\}}{\sqrt{[\zeta^2 + \gamma^2/4]} + \frac{\gamma}{2}} \right\}. \end{aligned} \quad (3.5)$$

This result gives

$$\frac{p(\rho)}{\rho kT} = -\Gamma + \frac{1}{2}\{1 + \sqrt{1 + 4\Gamma^2}\}. \quad (3.6)$$

We may calculate the grand canonical distribution functions as

$$\rho_{(++)}(x_A, x_B) = \Xi^{-1} g(x_A, \zeta, \gamma) g(x_B - x_B, \zeta, \gamma) f(L - x_B, \zeta, \gamma), \quad (3.7)$$

$$\rho_{(+-)}(x_A, x_B) = \Xi^{-1} g(x_A, \zeta, \gamma) f(x_B - x_B, \zeta, \gamma) k(L - x_B, \zeta, \gamma) \quad (3.8)$$

$$\rho_{(-+)}(x_A, x_B) = \Xi^{-1} f(x_A, \zeta, \gamma) h(x_B - x_A, \zeta, \gamma) f(L - x_B, \zeta, \gamma) \quad (3.9)$$

and

$$\rho_{(--)}(x_A, x_B) = \Xi^{-1} f(x_A, \zeta, \gamma) g(x_B - x_A, \zeta, \gamma) k(L - x_B, \zeta, \gamma). \quad (3.10)$$

In these expressions,

$$f(x, \zeta, \gamma) = \sum_{N=0}^{\infty} \zeta^{2N} F(x, N, \gamma) \quad (3.11)$$

where  $F$  is defined in equation (3.1) while

$$g(x, \zeta, \gamma) = \sum_{N=0}^{\infty} \zeta^{2N+2} G(x, N, \gamma) \quad (3.12)$$

$$h(x, \zeta, \gamma) = \sum_{N=0}^{\infty} \zeta^{2N+2} H(x, N, \gamma) \quad (3.13)$$

and

$$k(x, \zeta, \gamma) = \sum_{N=0}^{\infty} \zeta^{2N+2} K(x, N, \gamma) \quad (3.14)$$

with

$$G(x, N, \gamma) = \int_0^x dx_{2N+1} \int_0^{x_{2N+1}} dx_{2N} \int_0^{x_{2N}} dx_{2N-1} \cdots \int_0^{x_2} dx_1 \\ \times \exp \left( - \sum_{j=1}^{2N+1} \gamma(-1)^j x_j \right) \exp(-\gamma x) \quad (3.13)$$

$$H(x, N, \gamma) = \int_0^x dx_{2N-1} \int_0^{x_{2N-1}} dx_{2N-2} \cdots \int_0^{x_1} dx_0 \\ \times \exp \left( - \sum_{j=1}^{2N-1} \gamma(-1)^j x_j - \gamma x_0 - \gamma x \right). \quad (3.14)$$

and

$$K(x, N, \gamma) = \int_0^x dx_{2N} \int_0^{x_{2N}} dx_{2N-1} \int_0^{x_{2N-1}} dx_{2N-2} \cdots \int_0^{x_1} dx_0 \\ \times \exp \left( - \sum_{j=1}^{2N} \gamma(-1)^j x_j - \gamma x_0 - \gamma x \right). \quad (3.15)$$

The correlation functions in the bulk interior of the system can then be found by evaluating  $f$ ,  $g$ ,  $h$  and  $k$  by the same Laplace transform technique as was used for evaluating the grand canonical partition function. We obtain

$$f(x, \zeta, \gamma) = \frac{\zeta^2 e^{-\gamma x/2}}{2\sqrt{[\zeta^2 + \gamma^2/4]}} \left( \frac{e^{x\sqrt{[\zeta^2 + \gamma^2/4]}}}{\sqrt{[\zeta^2 + \gamma^2/4] - \gamma/2}} + \frac{e^{-x\sqrt{[\zeta^2 + \gamma^2/4]}}}{\sqrt{[\zeta^2 + \gamma^2/4] + \gamma/2}} \right), \quad (3.16)$$

$$g(x, \zeta, \gamma) = \frac{\zeta^2 e^{-\gamma x/2}}{2\sqrt{[\zeta^2 + \gamma^2/4]}} (e^{x\sqrt{[\zeta^2 + \gamma^2/4]}} - e^{-x\sqrt{[\zeta^2 + \gamma^2/4]}}), \quad (3.17)$$

$$h(x, \zeta, \gamma) = \frac{\zeta^4 e^{-\gamma x/2}}{2\sqrt{[\zeta^2 + \gamma^2/4]}} \left( \frac{e^{x\sqrt{[\zeta^2 + \gamma^2/4]}}}{\sqrt{[\zeta^2 + \gamma^2/4] + \gamma/2}} + \frac{e^{-x\sqrt{[\zeta^2 + \gamma^2/4]}}}{\sqrt{[\zeta^2 + \gamma^2/4] - \gamma/2}} \right), \quad (3.18)$$

and

$$k(x, \zeta, \gamma) = \frac{\zeta^2 e^{-\gamma x/2}}{2\sqrt{[\zeta^2 + \gamma^2/4]}} (e^{x\sqrt{[\zeta^2 + \gamma^2/4]}} - e^{-x\sqrt{[\zeta^2 + \gamma^2/4]}}). \quad (3.19)$$

We have

$$\rho_{(++)}(x_A, x_B) = \frac{1}{4}\rho^2 \{1 - \exp[-\rho\Delta x(1 + \sqrt{(1 + 4\Gamma^2)})]\} \quad (3.20)$$

where  $\Delta x = x_B - x_A$ . We have  $\rho_{(++)}(x_A, x_B) = \rho_{(--)}(x_A, x_B)$ . We also have

$$\rho_{(+-)}(x_A, x_B) = \frac{1}{4}\rho^2 \{1 + q(\Gamma) \exp[-\rho\Delta x(1 + \sqrt{(1 + 4\Gamma^2)})]\} \quad (3.21)$$

where

$$q(\Gamma) = \frac{1 + \sqrt{(1 + 4\Gamma^2)} - 2\Gamma}{1 + \sqrt{(1 + 4\Gamma^2)} + 2\Gamma} \quad (3.22)$$



and

$$\rho_{(-+)}(x_A, x_B) = \frac{1}{4}\rho^2 \left\{ 1 + \frac{1}{q(\Gamma)} \exp[-\rho \Delta x (1 + \sqrt{1 + 4\Gamma^2})] \right\}. \quad (3.23)$$

Notice that the  $(+ -)$  and  $(- +)$  distribution functions are not symmetric, an effect caused by the charge ordering on the line, among other things.

We may now turn to consider the fixed polarization ensemble. At fixed polarisation  $\sigma = M/L$ , the canonical partition function is

$$\mathbb{Z}_{2N}(L, \gamma, \sigma) = \int_0^L dx_{2N} \cdots \int_0^{x_2} dx_1 \exp(-\gamma M) \delta(M - L\sigma). \quad (3.24)$$

If we use the usual representation of the delta function as the inverse transform of 1 we obtain

$$\mathbb{Z}_{2N}(L, \gamma, \sigma) = \frac{1}{2\pi} \int_{-\infty}^{\infty} dk e^{-ikL\sigma} \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} \frac{ds}{s} e^{sL} \{s(s + \gamma - ik)\}^{-N}. \quad (3.25)$$

We may perform the integration over  $k$  exactly as a contour integral in the complex  $k$ -plane, closing the contour in the lower half plane and picking up the contribution of an  $N$ th order pole at  $k = -i(s + \gamma)$ , a number with positive real part. The resulting partition function is

$$\mathbb{Z}_{N(-+)}(L, \gamma, \sigma) = \frac{L^{N-1}}{(N-1)!} \sigma^{N-1} e^{-L\gamma\sigma} \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} ds s^{-(N+1)} e^{Ls(1-\sigma)}. \quad (3.26)$$

The integral over  $s$  may then be evaluated at large  $L$  by steepest descent. The free energy per unit length of system at fixed polarisation  $\sigma$  for a system charge ordered as  $- + - + \cdots - +$  is

$$\frac{f_{(-+)}(\rho, \sigma, \gamma)}{kT} = \rho \{ 2\Gamma\sigma - 1 - \frac{1}{2} \log [2\sigma(1-\sigma)] \} + \rho \log [\rho/\sqrt{2}]. \quad (3.27)$$

In the considerations of Section VII it will also be useful to consider the free energy density at fixed polarisation for systems which are ordered  $- + - + \cdots - + -$ ,  $+ - + - \cdots + -$  and  $+ - + - \cdots + - +$ . We may note at once that two of these cases are not exactly charge neutral, but nonetheless these thermodynamic free energy densities exist. The methods we use are the same as used on the problems above. For the  $+ - \cdots + -$  chain, the calculation is the same as the one already done, but with  $-\gamma$  replacing  $\gamma$  and  $-k$  replacing  $k$ . The resulting free energy density is

$$\frac{1}{kT} f_{(+ -)}(\rho, \sigma, \gamma) = \rho \{ 2\Gamma\sigma - 1 - \frac{1}{2} \log [2|\sigma|(1-|\sigma|)] \} + \rho \log [\rho/\sqrt{2}] \quad (3.28)$$

for  $\sigma < 0$  while  $f_{(+ -)}(\rho, \sigma, \gamma)$  diverges to  $+\infty$  for  $\sigma > 0$ . The calculations for the other two chains are very similar and give

$$f_{(++ )}(\rho, \sigma, \gamma) = f_{(-+ )}(\rho, \sigma, \gamma) \quad \text{and} \quad f_{(-- )}(\rho, \sigma, \gamma) = f_{(+ -)}(\rho, \sigma, \gamma).$$



A last free energy we shall require is that calculated in an ensemble where the polarisation density is constrained to lie between  $\sigma - \Delta/2$  and  $\sigma + \Delta/2$ . The partition function is

$$\mathbb{Z}_{2N}(L, \gamma, \sigma, \Delta) = \int_0^L dx_{2N} \cdots \int_0^{x_2} dx_1 \exp(-\gamma M) H(M, L\sigma, \Delta) \quad (3.29)$$

where

$$H(M, M', \Delta) = \begin{cases} 1 & \text{if } M' - L\Delta/2 < M < M' + L\Delta/2 \\ 0 & \text{otherwise.} \end{cases} \quad (3.30)$$

We can evaluate the partition function with this constraint using the Fourier transform representation for  $H$ . We find

$$\begin{aligned} \mathbb{Z}_{2N}(L, \gamma, \sigma, \Delta) &= \frac{1}{2\pi} \int_{-\infty}^{\infty} \frac{dk}{k} 2 \sin(kL\Delta/2) e^{-ikL\sigma} \\ &\quad \cdot \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} \frac{ds}{s} e^{sL} \{s(s + \gamma - ik)\}^{-N}. \end{aligned} \quad (3.31)$$

The evaluation proceeds by first doing the integral over  $k$  by steepest descent and then the integral over  $s$  by steepest descent. We may define the resulting free energy density as  $\Phi_{(-+)}(\rho, \sigma, \gamma, \Delta)$ . Its details are quite unimportant. It has the crucial property that

$$\lim_{\Delta \rightarrow 0} \Phi_{(-+)}(\rho, \sigma, \gamma, \Delta) = f_{(-+)}(\rho, \sigma, \gamma). \quad (3.32)$$

Similar results hold for the free energy densities for each of the other orderings of charges on the line.

#### IV. Periodic boundary conditions

In this section we use the Hamiltonian of equation (2.8),

$$\beta \mathcal{H}_{PBC} = -\frac{\gamma}{L} \left(M - \frac{L}{2}\right)^2 + \gamma L/4. \quad (2.8)$$

and the identity

$$\exp(\alpha x^2) = (\pi\alpha)^{-1/2} \int_{-\infty}^{\infty} \exp\left(-\frac{u^2}{\alpha} + 2ux\right) du. \quad (4.1)$$

These give

$$\mathbb{Z}_{2N}(L, N, \gamma) = \left(\frac{L}{\gamma\pi}\right)^{1/2} e^{-\gamma L/4} \int_{-\infty}^{\infty} e^{-Lu^2/\gamma - uL} F(L, N, -2u) \quad (4.2)$$

We may use the Laplace transform method, together with equation (3.3) to obtain the result

$$\mathbb{Z}_{2N}(L, N, \gamma) = \left(\frac{L}{\gamma\pi}\right)^{1/2} e^{-\gamma L/4} \int_{-\infty}^{\infty} e^{-Lu^2/\gamma - uL} du \cdot \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} \frac{e^{sL}}{s} \{s(s-2u)\}^{-N} ds \quad (4.3)$$

which gives for the thermodynamic limit of the free energy per particle

$$\frac{a(\rho, T)}{kT} - \{\log(\rho)/\sqrt{2}\} = \begin{cases} -\frac{1}{2}\{2 - \Gamma - \log(2)\} & \text{for } \Gamma < 1 \\ \frac{1}{2}\{-1 + \log(2\Gamma)\} & \text{for } \Gamma > 1. \end{cases} \quad (4.4)$$

We may calculate the grand canonical partition function immediately from equation (4.3). We obtain

$$\Xi(L, \xi, \gamma) = \left(\frac{L}{\gamma\pi}\right)^{1/2} e^{-\gamma L/4} \int_{-\infty}^{\infty} e^{-Lu^2/\gamma - uL} du \cdot \frac{1}{2\pi i} \int_{c-i\infty}^{c+i\infty} ds \frac{e^{sL}}{s} \frac{s(s-2u)}{s^2 - 2us - \xi^2}. \quad (4.5)$$

The inverse Laplace transform may be calculated exactly. Its exact form is not of much help since we must integrate it over  $u$ . The asymptotic behaviour, at large  $L$  is easy to obtain, and will suffice in the rest of this paper. We obtain

$$\Xi(L, \xi, \gamma) = \frac{1}{2} \xi^2 \left(\frac{L}{\gamma\pi}\right)^{1/2} e^{-\gamma L/4} \int_{-\infty}^{\infty} \frac{\exp\left(-L\left[\frac{u^2}{\gamma} - \sqrt{u^2 + \xi^2}\right]\right)}{u^2 + \xi^2 + u\sqrt{u^2 + \xi^2}} du. \quad (4.6)$$

A steepest descent estimate of this integral involves saddle points at  $u = 0$  and at  $\sqrt{u^2 + \xi^2} = \gamma/2$ . The second saddle points only occur for  $\xi \leq \gamma/2$  and in that circumstance they dominate the integral. To illustrate the behaviour of the saddle points, we write the exponential in the integrand in (4.6) as  $\exp[\gamma L \vartheta(u/\gamma)]$  and plot the function

$$\vartheta(v) = \sqrt{(\xi^2/\gamma^2 + v^2)} - v^2 \quad (4.7)$$

in Fig. 1, for  $\xi/\gamma = 0.3$  and  $0.7$ . There are two maxima on the curve for  $\xi/\gamma < 0.5$  and one for  $\xi/\gamma > 0.5$ .

The scaled pressure is readily found to be

$$\frac{p(\rho, t)}{\rho kT} = \begin{cases} 1 - \Gamma/2 & \text{for } 0 \leq \Gamma \leq 1 \\ \frac{1}{2} & \text{for } 1 \leq \Gamma \end{cases} \quad (4.8)$$

We plot this scaled pressure and the reduced free energy for the free boundary condition and periodic boundary condition cases in Figs 2 and 3. There is a phase

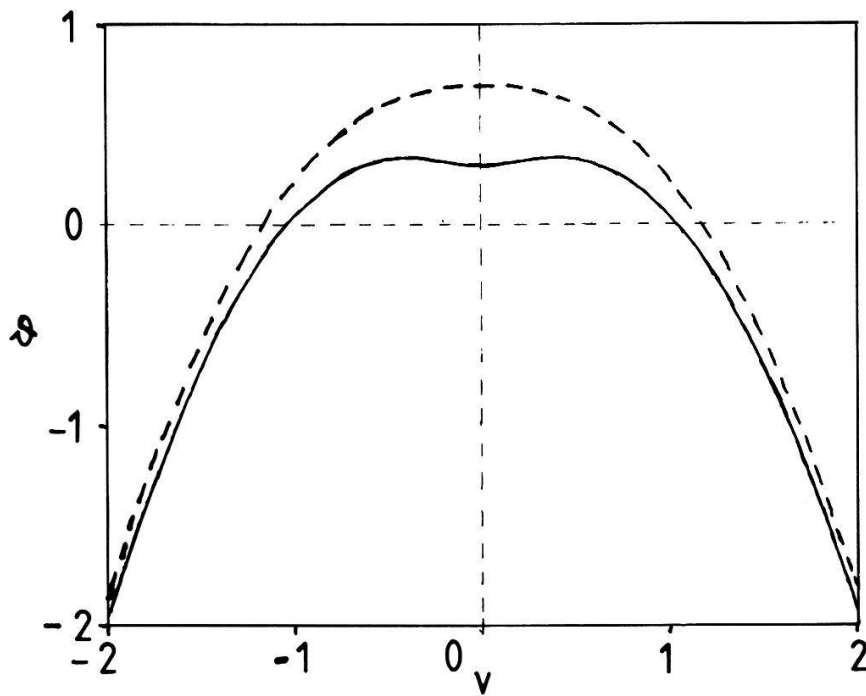


Figure 1  
Scaled exponent function  $\vartheta(v)$  for saddle point integration. Upper curve,  $\xi/\gamma = 0.7$ , lower curve,  $\xi/\gamma = 0.3$ .

transition in the periodic system, but not in the free boundary condition system. We defer discussion of the nature of this transition and the symmetry broken in it until the next section.

We now turn to discuss the distribution functions in the bulk interior of the system. We illustrate the method we use on the grand canonical distribution function  $\rho_{(++)}(x)$ , the distribution of pairs of positive particles in the bulk interior

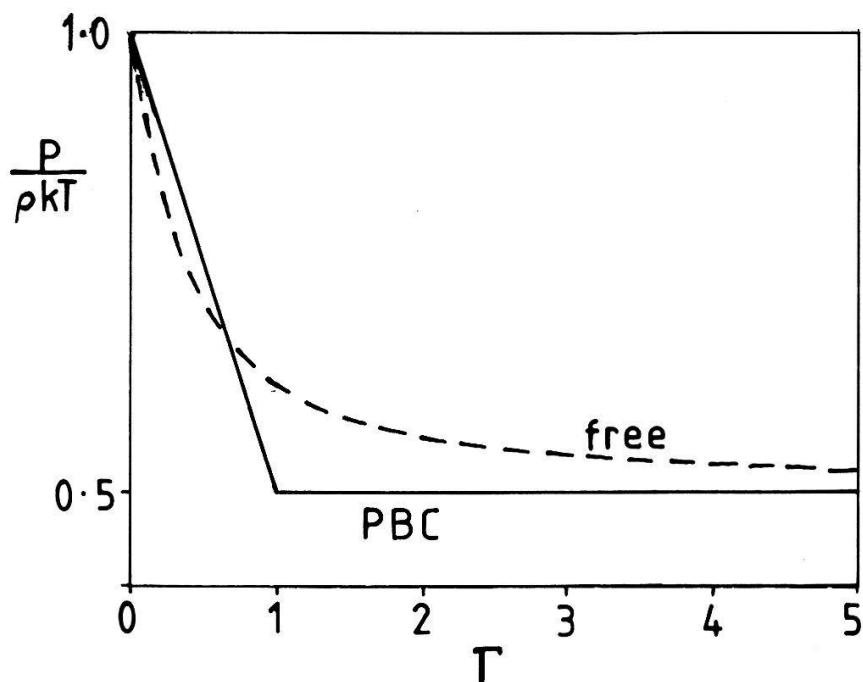


Figure 2  
One dimensional plasma pressures

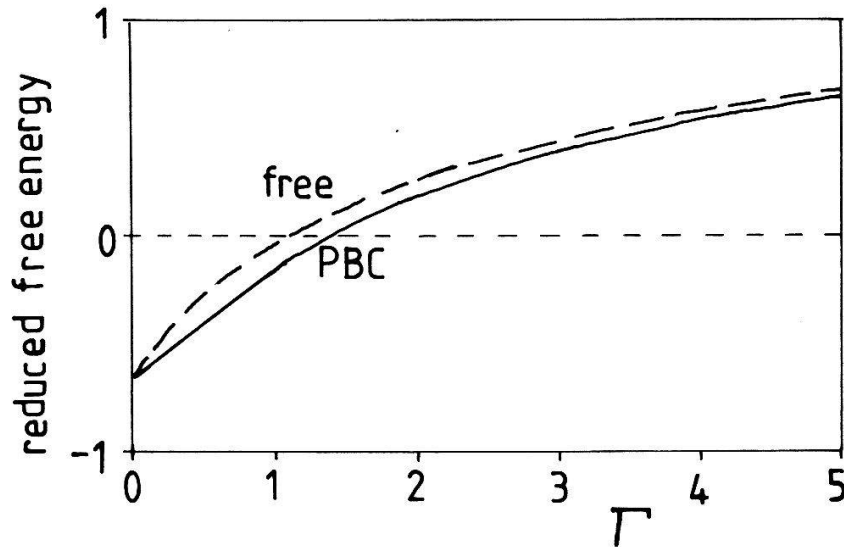


Figure 3  
Reduced free energies

of the system. We may write

$$\rho_{(++)}(x) = \Xi^{-1}(L, \zeta, \gamma) \left( \frac{L}{\gamma\pi} \right)^{1/2} e^{-\gamma L/4} \int_{-\infty}^{\infty} e^{-Lu^2/\gamma - uL} du \\ \times g(x_A, \zeta, -2u) g(x, \zeta, -2u) f(L - x_A - x, \zeta, -2u) \quad (4.9)$$

and this gives, in the limit,

$$\rho_{(++)}(x) = \begin{cases} \frac{1}{4}\rho^2\{1 - e^{-2x\rho}\} & \text{if } 0 \leq \Gamma \leq 1 \\ \frac{1}{4}\rho^2\{1 - e^{-2\Gamma x\rho}\} & \text{if } \Gamma \geq 1. \end{cases} \quad (4.10)$$

Exactly the same expression holds for  $\rho_{(--)}(x)$ . The other distribution functions are more complicated for the two saddle points contribute equally. If we assume that for  $\zeta \leq \gamma/2$  the saddle point at positive  $u$  (given by  $\sqrt{u^2 + \zeta^2} = \gamma/2$ ) dominates, then we have

$$\rho_{(+-)}(x) = \begin{cases} \frac{1}{4}\rho^2\{1 + e^{-2x\rho}\} & \text{for } 0 \leq \Gamma \leq 1 \\ \frac{1}{4}\rho^2\{1 + p(\Gamma)e^{-2x\rho\Gamma}\} & \text{for } \Gamma \geq 1 \end{cases}, \quad (4.11)$$

with

$$p(\Gamma) = \frac{1 + \sqrt{1 - 1/\Gamma}}{1 - \sqrt{1 - 1/\Gamma}} \quad (4.12)$$

and

$$\rho_{(-+)}(x) = \begin{cases} \frac{1}{4}\rho^2\{1 + e^{-2x\rho}\} & \text{for } 0 \leq \Gamma \leq 1 \\ \frac{1}{4}\rho^2\left\{1 + \frac{1}{p(\Gamma)}e^{-2x\rho\Gamma}\right\} & \text{for } \Gamma \geq 1 \end{cases}. \quad (4.13)$$

We again note that these distribution functions are nonsymmetric.

## V. The nature of the phase transition

Consider first the free boundary condition case. We evaluate the net charge clustered about a single fixed positive charge in the centre of the system in the thermodynamic limit. This charge is

$$Q_{\text{free}} = \frac{\rho}{2} \int_0^\infty [\rho_{(+, +)}(x) - \rho_{(+, -)}(x) + \rho_{(-, +)}(x) - \rho_{(-, -)}(x)] dx. \quad (5.1)$$

Using equations (3.20) to (3.23) we find  $Q_{\text{free}} = -1$ . This means that in spite of the asymmetry in the  $(+ -)$  and  $(- +)$  distribution functions, the system is locally neutral. We find the same result, namely  $Q_{PBC} = -1$ , for both  $\Gamma \leq 1$  and  $\Gamma \geq 1$  in the periodic boundary condition case.

We may now consider the mean polarisation of the system. We define this as

$$\mu(\Gamma) = \lim_{N \rightarrow \infty} \langle M \rangle / N = \lim_{N \rightarrow \infty} \frac{1}{N} \sum_{j=1}^N \langle x_{2j} - x_{2j-1} \rangle. \quad (5.2)$$

This may be readily calculated in the free boundary condition case as

$$\mu(\Gamma) = 2\{1 + 2\Gamma + \sqrt{1 + 4\Gamma^2}\}^{-1} \quad (5.3)$$

which lies on the interval  $[0, 1]$  for all real positive  $\Gamma$ .

In the periodic boundary condition case, things are not so simple, since we have two saddle points which can contribute at high coupling, each corresponding to a separate state of the system. We consider the effect of an infinitesimal electric field  $E$  on the system. This adds a term  $-QEM$  to the Hamiltonian. The appropriate scaled field parameter is  $\lambda = QE/kT$ . In this periodic boundary condition case, the functions  $f$ ,  $g$ ,  $h$  and  $k$  in the expressions like equation (4.8) for the distribution functions have their third argument  $-2u$  replaced by  $-(2u - \lambda)$ . This means that in the saddle point estimations we obtain an exponent function of the form  $\exp(L\{\sqrt{[(u - \lambda/2)^2 + \xi^2]} - u^2/\gamma\})$ . Limitingly small positive fields thus select out the saddle point at  $u = -\sqrt{(\gamma^2/4 - \xi^2)}$  for  $\xi < \gamma/2$ . We may now examine the mean polarization  $\langle M \rangle$  in these boundary conditions. We define the order parameter

$$\tau = [\rho\mu(\Gamma) - 1]/2 = \sigma - \frac{1}{2}, \quad (5.2)$$

which is the deviation of the polarisation density from its value when the ordered particles are evenly spaced on the line.

In the free boundary condition case, in either sign of  $E$  we obtain

$$\tau_{\text{free}} = \frac{1}{2} \left( \frac{1 - 2\Gamma - \sqrt{1 + 4\Gamma^2}}{1 + 2\Gamma + \sqrt{1 + 4\Gamma^2}} \right) \quad (5.3)$$

which is always less than zero. If the particles are evenly spaced,  $\sigma = 1$ . Thus in free boundary conditions, the particles are always bound into dipolar molecules, reflecting much of what is known of one dimensional coulombic systems.

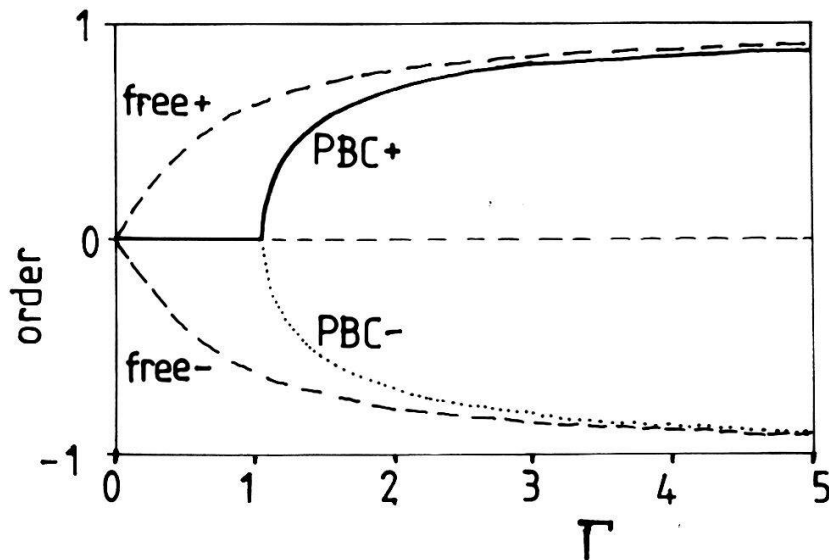


Figure 4  
Order parameter for one dimensional plasma

In the periodic boundary condition case, we obtain

$$\tau_{PBC} = \begin{cases} 0 & \text{for } 0 \leq \Gamma \leq 1 \\ \pm[1 - 1/\Gamma]^{1/2} & \text{for } \Gamma \geq 1. \end{cases} \quad (5.4)$$

We illustrate both order parameters in Fig. 4. Note that the free boundary condition system may have a positive  $\sigma$  curve if the particles are given the  $+ -$  alternating order on the line. Such a curve is naturally the reflection of the one sketched here. Consideration of the spacing which gives rise to the structure of these curves tells us the nature of the phases in the periodic boundary condition case. For  $\Gamma \leq 1$ , the order parameter is zero and the particles have a average spacing of  $1/\rho$ . This phase may be called a plasma phase because the charges are not bound together at all. For  $\Gamma \geq 1$ , in the phase selected as  $E \rightarrow 0^+$ , we have  $\sigma_{PBC} < 0$ . In this phase the charged particles cluster in close  $(- +)$  pairs along the line. We may call this phase dipolar, because of the clustering. For  $\Gamma \geq 1$ , in the phase selected as  $E \rightarrow 0^-$ , we have  $\sigma_{PBC} > 0$ . In this phase the nearest neighbour  $(- +)$  pairs separate so that there are close  $(+ -)$  pairs along the line. This of course leaves two charges unpaired at either end of the system. Those two charges may be seen as paired with particles from the next periodic image of the system. One of the reasons we do not see such behaviour in the free boundary condition system is that no such pairing of end particles is available.

For the system of evenly spaced particles,  $\tau = 0$ , so that the phase transition is from a plasma state to a bound pair state with a polarisation per bound pair equal to  $1 \pm \tau$ . The critical point of the transition is classical. We return to that point for a moment in the next section.

## VI. Zeros of the grand canonical partition function

The system we have discussed here is interesting not just because it is a Coulombic system but also because we can solve it exactly. We can make useful

statements about the zeros of the grand canonical partition function as a function of its natural variable  $\zeta^2$  in both the free boundary condition case and the periodic boundary condition case.

In the free boundary condition case the grand canonical partition function may be written in the form

$$\Xi(L, \zeta, \gamma) = e^{-\alpha} \left\{ \cosh(\xi) + \frac{\alpha}{\xi} \sinh(\xi) \right\}. \quad (6.1)$$

Here  $\alpha = \gamma L/2 = 2N\Gamma$  and  $\xi = L\sqrt{(\zeta^2 + \gamma^2/4)} = 2N((\zeta/\rho)^2 + \Gamma^2)^{1/2}$ . The zeros of this partition function as a function of  $\xi$  are at the non-zero solutions of  $\xi = -\alpha \tanh(\xi)$ , and these are confined to the imaginary  $\xi$  axis. For  $\xi = i\phi$  and  $(n - \frac{1}{2})\pi < \phi < n\pi$ , with  $n > 0$ , we have the equation  $\tan(\phi) = -\alpha\phi$ . The right hand side of this equation is negative, decreasing monotonically and continuous on the subinterval we are considering, while the left hand side is monotonic increasing and spans from  $-\infty$  to 0. There is then exactly one zero  $\phi_n$ ,  $n = \pm 1, \pm 2, \dots$  in each interval  $(n - \frac{1}{2})\pi < \phi < n\pi$ . The zeros of the partition function are thus on the negative real axis of the complex  $\zeta^2$  plane. In the thermodynamic limit we have the density of zeros

$$\Sigma(x) = \frac{1}{2\pi} [-x - \gamma^2/4]^{-1/2} \quad (6.2)$$

on the segment of this axis with  $\zeta^2 < -\gamma^2/4$ . We can reconstruct the pressure from this zero density in the usual way.

For the periodic boundary condition case we have not found such an explicit description of all the partition function zeros. However, we can find an asymptotic representation for the grand canonical partition function near  $\zeta^2 = \gamma^2/4$  from equation (4.6). If we take the integral in that equation and change variable to  $u = v(L/\gamma^3)^{1/4}$  and introduce  $y = 2(L/\gamma^3)^{1/2}[\zeta^2 - \gamma^2/4]$  and  $\psi = L\gamma$ , then we find

$$\Xi(L, \zeta, \gamma) = \frac{1}{\sqrt{\pi}} \{ \psi \exp[\psi(1 + 2y)] \}^{1/4} \int_0^\infty dv \exp(-yv^2 - v^4) \{1 + O(L^{-1/2})\}. \quad (6.3)$$

The integral here has been investigated by Glasser et al. [12] in studies of the complex temperature plane zeros for partition functions of mean field models of Ising models. They show that

$$\int_0^\infty dv \exp(-yv^2 - v^4) = \frac{1}{4} y^{1/2} \exp(y^2/8) K_{1/4}(y^2/8) \quad (6.4)$$

where  $K_\nu(z)$  is the modified Bessel function of the second kind of order  $\nu$ . They appeal to Watson [13] to show that there is an infinite series of zeros of this function located asymptotically at

$$y_n = (4\pi n)^{1/2}(-1 + i)\{1 + O(1/n)\}, \quad n = 1, 2, 3, \dots \quad (6.5)$$



together with another conjugate set at  $y_n^*$ . The zeros accumulate on lines intersecting the positive  $\zeta^2$  axis at  $\zeta^2 = \gamma^2/4$  at an angle of  $3\pi/4$ . The normal Yang–Lee picture for the zeros of the grand partition function as a function of fugacity at a classical phase transition is that the zeros should accumulate on a line which cuts the fugacity axis at an angle of  $\pi/2$ . This picture was confirmed by Hemmer et al. [14–15]. However, the symmetry breaking field for this transition is an electric field, so it is not surprising that the fugacity behaves as a temperature like variable at the transition. This picture of the zeros is unfortunately incomplete: we cannot see how the zero distribution is perturbed by the periodic boundary conditions, at least not in any easily apparent analytical way. To uncover their behaviour would require detailed numerical calculation.

## VII. Perturbation theory for the periodic boundary conditions

We consider now a system of  $2N$  charges on a line of length  $L$  in periodic boundary conditions. We divide the line into  $P$  sub intervals of length  $l$  so that  $Pl = L$ . We consider contributions to the canonical partition function in which the  $\lambda$ th sub interval contains  $N_\lambda$  particles and has a mean polarisation  $\sigma(\lambda) - \Delta/2 < \langle \sigma \rangle < \sigma(\lambda) + \Delta/2$ . The values of  $\langle \sigma \rangle$  for the sub intervals must lie on  $-1 \leq \langle \sigma \rangle \leq 1$  so we allow the  $\sigma(\lambda)$  to have the values  $n/N$ ,  $-N \leq n \leq N$ , with  $N$  being the integer part of  $2/\Delta$ . There are  $(L\rho + 1)^P$  ways of distributing the particles over the subintervals and for each of these there are  $(2/\Delta + 1)^P$  ways of choosing the  $\sigma(\lambda)$ ,  $\lambda = 1, 2, \dots, P$ . A particular choice of  $\{N_\lambda, \sigma(\lambda), 1 \leq \lambda \leq P\}$  gives a contribution to the partition function. We define

$$\sigma_-(\lambda) = \text{sgn}(\sigma(\lambda))\{|\sigma(\lambda)| - \Delta/2\} \quad (7.1a)$$

and

$$\sigma_+(\lambda) = \text{sgn}(\sigma(\lambda))\{|\sigma(\lambda)| + \Delta/2\}. \quad (7.1b)$$

We now make upper and lower bounds on the partition function, remembering that the Hamiltonian is

$$-\beta\mathcal{H}_{PBC} = -\gamma \sum_{\lambda=1}^P M(\lambda) + \frac{\gamma}{L} \left( \sum_{\lambda=1}^P M(\lambda) \right)^2 \quad (7.2)$$

where for the  $N_\lambda$  particles on subinterval  $\lambda$ ,

$$M(\lambda) = \sum_{j=1}^{N_\lambda} e_j x_j. \quad (7.3)$$

We then have

$$\begin{aligned}
 & \mathbb{Z}_{2N}(L, \gamma; PBC) \\
 &= \sum_{\{N_\lambda\}} \sum_{\{\sigma(\lambda)\}} \int_0^l dx_{N_1} \cdots \int_0^{x_2} dx_1 e^{-\gamma M(1)} H(M(1), l\sigma(1), \Delta) \\
 & \quad \times \int_0^l dx_{N_2} \cdots \int_0^{x_2} dx_1 e^{-\gamma M(2)} H(M(2), l\sigma(2), \Delta) \\
 & \quad \dots \\
 & \quad \times \int_0^l dx_{N_P} \cdots \int_0^{x_2} dx_1 e^{-\gamma M(P)} H(M(P), l\sigma(P), \Delta) \\
 & \quad \times \exp \left\{ \frac{\gamma}{L} \left( \sum_{\lambda=1}^P M(\lambda) \right)^2 \right\}. \tag{7.4}
 \end{aligned}$$

We can make a lower bound on the partition function by replacing the square of the sum of the  $M(\lambda)$  in the exponential at the end of this integral by the square of the sum of the  $\sigma_-(\lambda)$  and taking the largest term in the sum on  $\{N_\lambda\}$  and  $\sigma(\lambda)\}$ . We may make an upper bound on the partition function by using  $\sigma_+(\lambda)$  and the largest resulting term times the number of terms in the sum. We then take the thermodynamic limit by writing out the double inequality that results, taking the log of each term in it, which maintains the inequality, dividing by  $L$ , letting  $l \rightarrow \infty$  at fixed  $P$ , then letting  $P \rightarrow \infty$  and finally letting  $\Delta \rightarrow 0$ . We use the constant polarisation free energy densities introduced in Section III and shown there to exist by explicit calculation.

A problem arises with those cells which contain an odd number of particles. Consider the cell between  $\nu l$  and  $(\nu + 1)l$ . If it has particles in the  $(- -)$  configuration, the free energy density contains an extra term  $-2\Gamma\rho\nu$  and if in the  $(+ +)$  configuration the free energy density contains an extra term  $2\Gamma\rho\nu$ . When we take account of the ordering of such configurations forced by the overall charge ordering and the extra dipole moments of non-neutral cells, we find

- (i) a  $(- +)$  cell contributes  $f_{(-+)}(\rho, \sigma, \gamma)/P$  to the free energy,
- (ii) a  $(+ -)$  cell contributes  $f_{(-+)}(\rho, 1 - |\sigma|, \gamma)/P$  to the free energy,
- (iii) a  $(- -)$  cell contributes  $f_{(-+)}(\rho, 1 - |\sigma|, \gamma)/P$  to the free energy, and
- (iv) a  $(+ +)$  cell contributes  $f_{(-+)}(\rho, \sigma, \gamma)/P$  to the free energy.

By taking the limiting process described above we find that the free energy density in periodic boundary conditions is

$$\begin{aligned}
 \frac{f_{PBC}(\rho, \gamma)}{kT} = \min_{\rho \in \mathcal{C}_1} \min_{\sigma \in \mathcal{C}_2} & \left( \int_0^1 dx \rho(x) \left\{ \log [\rho(x)/\sqrt{2}] \right. \right. \\
 & \left. \left. + 2\Gamma\sigma(x) \left[ 1 - \int_0^1 \sigma(y) dy \right] - 1 - \frac{1}{2} \log [2\sigma(x)(1 - \sigma(x))] \right\} \right). \tag{7.5}
 \end{aligned}$$

In equation (7.5),  $\mathcal{C}_1$  is the class of functions  $[0, 1] \rightarrow \mathbb{R}$  with  $\rho(x) \geq 0$  and  $\int_0^1 \rho(x) dx = \rho$ . The class of functions  $\mathcal{C}_2$  is the class of polarisation density functions  $[0, 1] \rightarrow \mathbb{R}$  with  $0 \leq \sigma(x) \leq 1$ . This functional is minimised by a constant

density and a constant polarisation density, which is  $\frac{1}{2}$  for  $\Gamma \leq 1$  and may take two values for  $\Gamma \geq 1$ . We find

$$\sigma_{\min} = \begin{cases} \frac{1}{2} & \text{for } 0 \leq \Gamma \leq 1 \\ \frac{1}{2}\{1 + \sqrt{1 - 1/\Gamma}\} & \text{for } \Gamma \geq 1. \end{cases} \quad (7.6)$$

The free energy density at the minimum is given by

$$\frac{f(\rho, T)}{\rho kT} - \log[\rho/\sqrt{2}] = \begin{cases} -\frac{1}{2}\{2 - \Gamma - \log(2)\} & \text{for } 0 \leq \Gamma \leq 1 \\ \frac{1}{2}\{-1 + \log(2\Gamma)\} & \text{for } \Gamma \geq 1. \end{cases} \quad (7.7)$$

This is exactly the free energy density in periodic boundary conditions, as can be seen from equation (4.4)

We see that the extra potential introduced into the system by periodic boundary conditions can be treated by considering it to be a Kac potential and using the methods of Lebowitz–Penrose theory. These general methods have been augmented by the techniques of Millard and Leff [16, 17] to handle the problem which occurs with the Lebowitz–Penrose construction when the order parameter is based on a continuous microscopic variable like the polarisation density, rather than particle numbers as in the liquid gas case.

### VIII. Conclusions

The model introduced here is a Coulombic model: it displays exact local charge neutrality. The periodic boundary conditions much loved by simulators and presenters of exact solutions for statistical mechanical models produces a phase transition which is absent in free boundary conditions. That is a fairly significant change to be caused by boundary conditions!

Further, the rigorous perturbation theory developed in Section VII shows that the effect of the extra potential introduced by the periodic boundary conditions may be predicted by a variant of Lebowitz–Penrose theory for the effect of a weak long ranged potential. It may be noted that if we replace free boundary conditions by periodic boundary conditions for Coulombic systems in two or three dimensions, the potential shifts in exactly the same way. The  $1/|\mathbf{r}|$  potential has added to it a potential of the form  $(1/L)\psi(\mathbf{r}/L)$  (where  $L$  is the side of the cube of periodicity), and the precise nature of the function  $\psi$  depends on the nature of the boundary conditions used [18–20]. Part of the function  $\psi$  is a square of the dipole moment of the system, which makes the ideas developed in this paper apparently relevant.

It would appear then that to sort out the effects of periodic boundary conditions on the free energy of a Coulombic system it will be necessary to prove the existence of a free energy density at constant polarisation for free boundary conditions. With the existence of such a free energy density it may be possible to construct a Lebowitz–Penrose theory of the effects of periodic boundary conditions. The sorts of effects that periodic boundary conditions can have are not clear. If a system has a Hamiltonian which permits the development of a net

polarisation, then these extra terms from periodic boundary conditions will certainly affect the domain structure of the sample and may even affect the phase transition to the polarised state. Such systems do occur in nature and so we must be a little careful in the way we model the behaviour of materials like ferroelectric crystals.

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