

Interaction between two rows of localized adsorption sites in a 2D one-component plasma

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We compute the free energy for two rows of localized adsorption sites embedded in a two dimensional one-component plasma with neutralizing background density ρ . The interaction energy between the adsorption sites is repulsive. We also compute the average occupation number of the adsorption sites and compare it to the result for a single row of sites. The exact result indicates that the discretization does not induce charge asymmetry and no attractive forces occur.

Key words: *one-component plasma, localized adsorption, DNA attraction*

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1. Introduction

The subtleties of electrostatics in condensed matter theory represent a formidable and never ending challenge. One topic of much recent activity, has been the attraction between two macromolecules of the same charge [1]. One mechanism that has been proposed invokes charge asymmetry related to the formation of lattices or Wigner crystals [2,3]. One problem with this picture is that it will create a dipole that is inconsistent with the perfect screening sum rules (Blum et al.[4]). While the formation of Wigner crystals under special conditions is an experimental fact, the question of the large asymmetry in the charge distribution needs clarification. As has happened in the past the exact solution of the two dimensional Jancovici model [5] can provide an unambiguous answer to the puzzle. The interaction between two equally charged lines (which are charged surfaces in 2 dimensions!) has been

discussed using the exact solution, and is always repulsive [6]. Here, we study a discretized version of this problem, namely two lines of discrete adsorption sites, where the adsorption potential is given by the Baxter [7] sticky potential. To do this we extend the localized adsorption model of a single line [8] to the case of two lines of discrete adsorption sites. This extension is non-trivial, and as in a similar case discussed in the past, has a simple solution for what we would call a “commensurate” lattice [9], namely the spacing of the adsorption sites is such that the background charge of the enclosed area corresponds to an entire number of discrete charges.

2. Formalism

2.1. Modeling the adsorption

Following Rosinberg et al. [8], the adsorption potential for a sticky site located at the origin, given by $u_a(\mathbf{r})$, is modeled as

$$\exp[-\beta u_a(\mathbf{r})] = 1 + \lambda \delta(\mathbf{r}), \quad (1)$$

where λ is a positive constant that measures the strength of the adsorption potential [7].

The partition function for a system of adsorption sites with locations given by the vectors \mathbf{R}_m is given by

$$Z_N = \frac{1}{N!} \int e^{-\beta V_0(\mathbf{r}_1, \dots, \mathbf{r}_N)} \prod_{i=1}^N \left\{ \left[1 + \lambda \sum_{m=1}^M \delta(\mathbf{r}_i - \mathbf{R}_m) \right] d^2 r_i \right\}, \quad (2)$$

where V_0 is the potential energy of the one-component plasma in the absence of adsorption sites. Expanding in powers of λ , it has been shown [8] that the partition function can be written in terms of the n -point correlation functions as

$$Z_N = Z_N^0 \sum_s \sum_{m_1, m_2, \dots, m_s=1}^M \frac{\lambda^s}{s!} \rho(\mathbf{R}_{m_1}, \dots, \mathbf{R}_{m_s}), \quad (3)$$

where Z_N^0 is the partition function of the unperturbed system.

The difference in free energy from the unperturbed system is the logarithm of Z_N/Z_N^0 , and is given by

$$\Delta F = -k_B T \sum_{s \geq 1} \frac{\lambda^s}{s!} T_s, \quad (4)$$

where

$$T_s = \sum_{m_1, \dots, m_s=1}^M \rho_T(\mathbf{R}_{m_1}, \dots, \mathbf{R}_{m_s}), \quad (5)$$

and where ρ_T gives the truncated n -body correlation functions,

$$\begin{aligned} \rho_T(\mathbf{r}_1) &= \rho(\mathbf{r}_1), \\ \rho_T(\mathbf{r}_1, \mathbf{r}_2) &= \rho(\mathbf{r}_1, \mathbf{r}_2) - \rho(\mathbf{r}_1)\rho(\mathbf{r}_2), \\ \rho_T(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) &= \rho(\mathbf{r}_1, \mathbf{r}_2, \mathbf{r}_3) - \rho(\mathbf{r}_1, \mathbf{r}_2)\rho(\mathbf{r}_3) - \dots \end{aligned} \quad (6)$$

2.2. Correlation functions of the one-component plasma

The exact solution to the one-component plasma found by Jancovici [5] for coupling parameter $\Gamma = Z^2 e^2 / (k_B T) = 2$, where Z is the ion valence and e is the elementary charge of an electron. The n -point density correlation functions are given by

$$\rho(\mathbf{r}_1, \dots, \mathbf{r}_n) = \rho^n \det \left[e^{-\pi\rho(|z_\mu|^2 + |z_\gamma|^2)/2 + \pi\rho z_\mu \bar{z}_\gamma} \right] \Big|_{\mu, \gamma=1, \dots, n}. \quad (7)$$

Here, ρ is the background charge density, $z = x + iy$ where x and y describe coordinates on the plane, and \bar{z} its complex conjugate. After some algebraic manipulation, we can rewrite this expression as

$$\rho(\mathbf{r}_1, \dots, \mathbf{r}_n) = \rho^n \sum_{\sigma \in S_n} \text{sgn}(\sigma) \prod_{j=1}^n \rho_0(\mathbf{r}_j - \mathbf{r}_{\sigma(j)}) \rho_X(\mathbf{r}_j, \mathbf{r}_{\sigma(j)}), \quad (8)$$

where S_n is the group of permutations on n letters,

$$\rho_0(\mathbf{r}) = \exp\left(-\frac{\pi\rho}{2}\mathbf{r}^2\right), \quad (9)$$

and

$$\rho_X(\mathbf{r}, \mathbf{r}') = \exp[-i\pi\rho(\mathbf{r} \times \mathbf{r}') \cdot \hat{z}]. \quad (10)$$

The truncated correlation functions, $\rho_T(\mathbf{r}_1, \dots, \mathbf{r}_n)$ are computed by restricting the sum over σ in equation (8) to n -cycles.

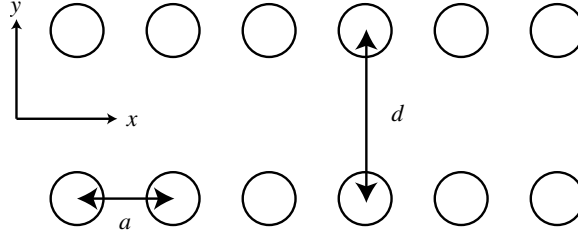


Figure 1. Two infinite lines of adsorption sites separated by a distance d and having periodicity a .

We suppose that the two lines of sticky sites with periodicity a are separated by a distance d (see figure 1). We describe the sticky site locations by introducing integer variables n_i and Ising variables δ_i that take on a value of either 0 or 1. Then any \mathbf{R}_i can be written as

$$\mathbf{R}_i = an_i \hat{x} + d\delta_i \hat{y}. \quad (11)$$

Since we are calculating the sums over all the positions of the particles, all n -cycles are equivalent by a suitable relabeling of the summation indices. This leads to the general expression

$$\begin{aligned} T_s = & \rho^s (s-1)! (-1)^s \sum_{n_1, \dots, \delta_1, \dots} \rho_0(\mathbf{R}_{m_1, \delta_1} - \mathbf{R}_{m_2, \delta_2}) \rho_0(\mathbf{R}_{m_2, \delta_2} - \mathbf{R}_{m_3, \delta_3}) \dots \\ & \times \rho_0(\mathbf{R}_{m_s, \delta_s} - \mathbf{R}_{m_1, \delta_1}) \rho_X(\mathbf{R}_{m_1, \delta_1}, \mathbf{R}_{m_2, \delta_2}) \rho_X(\mathbf{R}_{m_2, \delta_2}, \mathbf{R}_{m_3, \delta_3}) \dots \rho_X(\mathbf{R}_{m_s, \delta_s}, \mathbf{R}_{m_1, \delta_1}). \end{aligned} \quad (12)$$

3. Free energy

We first consider $\rho_X(\mathbf{R}_1, \mathbf{R}_2)$, and substitute in the adsorption site positions from equation (11). This gives

$$\rho_X(\mathbf{R}_1, \mathbf{R}_2) = e^{-i\pi\rho ad(n_1\delta_2 - \delta_1 n_2)}. \quad (13)$$

For the particular choice of background charge density

$$\rho = 2m/(ad), \quad (14)$$

where m is a positive integer, $\rho_X = 1$ when evaluated on the adsorption sites. We therefore specialize to densities where this simplification occurs.

We also find that $(\mathbf{R}_1 - \mathbf{R}_2)^2 = a^2(n_1 - n_2)^2 + d^2(\delta_1 - \delta_2)^2$, leading to

$$T_s = \rho^s (s-1)! (-1)^s \sum_{n_1, \dots} e^{-t[(n_1 - n_2)^2 + \dots + (n_s - n_1)^2]} \sum_{\delta_1, \dots} e^{-t'[(\delta_1 - \delta_2)^2 + \dots + (\delta_s - \delta_1)^2]}, \quad (15)$$

where $t = \pi\rho a^2/2$ and $t' = \pi\rho d^2/2$. Since the sum over the n_i and the sum over the δ_i decompose, we can evaluate the sum over δ_i using a transfer matrix. We define the transfer matrix, \mathcal{T} , to have the components

$$\mathcal{T} = \begin{pmatrix} 1 & e^{-\pi\rho d^2/2} \\ e^{-\pi\rho a^2/2} & 1 \end{pmatrix}. \quad (16)$$

Then

$$T_s = \rho^s (s-1)! (-1)^s \text{tr}(\mathcal{T}^s) \sum_{n_1, \dots} e^{-t[(n_1 - n_2)^2 + \dots + (n_s - n_1)^2]}. \quad (17)$$

Diagonalizing \mathcal{T} gives the eigenvalues $\lambda_{\pm} = 1 \pm e^{-\pi\rho d^2/2}$, allowing us to take the trace easily. Notice that the decoupling of the Ising variables, δ_i and integer variables, n_i only decouple at densities given by equation (14). At other densities, these additional couplings between the n_i and δ_i complicate the evaluation of the transfer matrix trace.

The sum over the n_i can be expressed in terms of Jacobi theta functions [8], where the Jacobi theta function is defined as

$$\theta_3(\zeta, t) = \sum_{n=-\infty}^{\infty} e^{-tn^2} e^{2i\pi n\zeta}. \quad (18)$$

First, notice that

$$\int d\zeta \theta_3^s(\zeta, t) = \sum_{n_1, \dots, n_{s-1}} e^{-t[n_1^2 + n_2^2 + \dots + (n_1 + n_2 + \dots)^2]} \quad (19)$$

for $s > 1$ and $\int d\zeta \theta_3(\zeta, t) = 1$. This leads to the expression

$$T_s = \rho^s (s-1)! (-1)^s \left[(1 + e^{-\pi\rho d^2/2})^s + (1 - e^{-\pi\rho d^2/2})^s \right] \int_0^1 d\zeta \theta_3(\zeta, \pi\rho a^2/2)^s, \quad (20)$$

Substituting this into equation (4) leads to a sum of two series, both of which are absolutely summable when $|\lambda\rho(1+e^{-\pi\rho d^2/2})\theta(0,t)| < 1$. By analytic continuation, we extend this sum to the full range of parameters, leading to the free energy difference between the OCP with and without adsorption sites given by

$$\Delta f = -\frac{k_B T}{a^2} \int_0^1 d\zeta \ln \left[\left\{ 1 + \lambda\rho e^{-(\pi\rho d^2/2)} \theta_3(\zeta, \pi\rho a^2/2) \right\}^2 - \lambda^2 \rho^2 e^{-\pi\rho d^2} \theta_3^2(\zeta, \pi\rho a^2/2) \right]. \quad (21)$$

In the limit that $d \rightarrow \infty$, we expect the free energy to be a sum of the free energies of two independent lines of sticky sites. Indeed, this limit yields

$$\Delta f_{d \rightarrow \infty} = -2 \frac{k_B T}{a^2} \int_0^1 d\zeta \ln [1 + \lambda\rho \theta_3(\zeta, \pi\rho a^2/2)]. \quad (22)$$

In the opposite limit, $d \rightarrow 0$, we expect the free energy to agree with that of a single line of adsorption sites with a potential given by 2λ . It is easy to see that the free energy in this limit is

$$\Delta f_{d \rightarrow 0} = -\frac{k_B T}{a^2} \int_0^1 d\zeta \ln [1 + 2\lambda\rho \theta_3(\zeta, \pi\rho a^2/2)]. \quad (23)$$

Written in terms of the dimensionless constants $t = \pi\rho a^2/2 = \pi a/d$, $t' = \pi\rho d^2/2 = \pi d/a$ and $\Lambda = \lambda\rho = 2\lambda/(ad)$, we find the change in free energy as the adsorption sites approach each other, $\Delta F = \Delta f - \Delta f_{d \rightarrow \infty}$, to be given by

$$\Delta F = -\frac{k_B T}{a^2} \int_0^1 d\zeta \ln \left[\frac{1 + \Lambda(1 + e^{-t'})\theta_3(\zeta, t)}{1 + \Lambda\theta_3(\zeta, t)} \right] - \frac{k_B T}{a^2} \int_0^1 d\zeta \ln \left[\frac{1 + \Lambda(1 - e^{-t'})\theta_3(\zeta, t)}{1 + \Lambda\theta_3(\zeta, t)} \right]. \quad (24)$$

Recall that this free energy is only valid when $\rho ad/2 = m$ for any positive integer m . Thus, we can compare the free energy of states with the same lattice constant a and background charge density ρ , only for integer multiples of some specific valid separation d . To be more specific, suppose we have the free energy at some density such that $\rho ad/2 = 1$ and separation d , then at separation md in the same background density, we have $\rho ad/2 = m$. Thus, the free energy formula equation (21) will be valid only for integer multiples of d .

In figure 2, we plot ΔF as a function of d for two values of the background density, ρ , given by $\rho ad_0/2 = 1$ and $\rho ad_0/2 = 5$, where $d_0 = 0.1a$ is the smallest value of d . The lines of adsorption sites are always repulsive. For larger d , ΔF becomes zero quickly. In figure 3, we plot ΔF as a function of d for different values of λa^2 . As the depth of the potential increases, the repulsive strength of the interaction also increases. This is indicative of the adsorption sites pinning charge on them, leading to their repulsion.

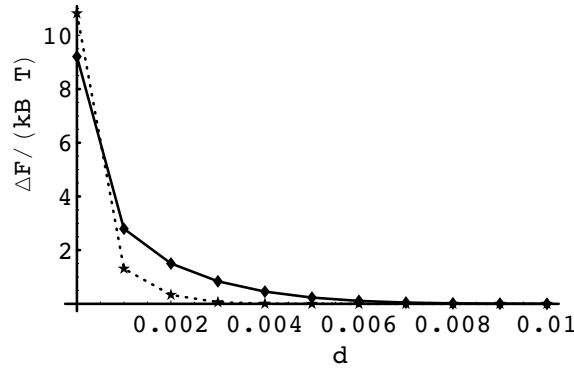


Figure 2. A typical set of free energies $\Delta f - \Delta f_{d \rightarrow \infty}$ with background density $\rho a d_0/2 = 1$ (circles, solid lines) and $\rho a d_0/2 = 5$ (stars, dotted lines). Here, d is measured in units of a and is an integer multiple of $0.1a$. We further set $\lambda a^2 = 100$. The lines are guides to the eye.

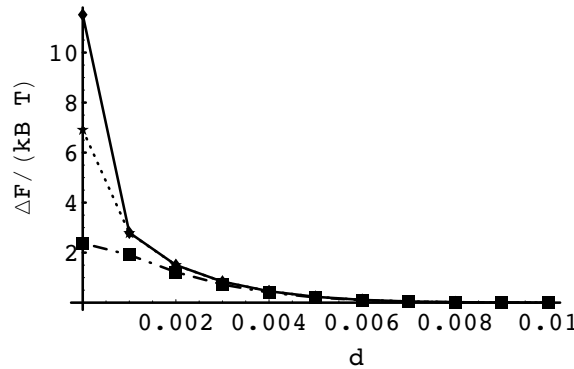


Figure 3. A typical set of free energies $\Delta f - \Delta f_{d \rightarrow \infty}$ with background density $\rho a d_0/2 = 1$ for values of $\lambda a^2 = 0.001$ (squares, dashed line), 0.1 (stars, dotted line), 10 (circles, solid line). Here, d is measured in units of a and is an integer multiple of $0.1a$. The lines are guides to the eye.

Finally, we compute the average occupation number of a site. This is given by [8]

$$\langle n \rangle = -a^2 \lambda \frac{\partial}{\partial \lambda} \frac{\Delta f}{k_B T}. \quad (25)$$

This is plotted as a function of λa^2 for in figure 4 for two lines with separation $d = 0.1a$ and $\rho a d/2 = 1$ (solid line) and for a single line of adsorption sites (dashed line).

It is clear from figure 4 that the repulsion inhibits the adsorption of the ions. However, the separation $d = 0.1a$ is very small. At separations on the order of the site spacing, there is no appreciable difference in the fraction of occupied sites as a function of λ .

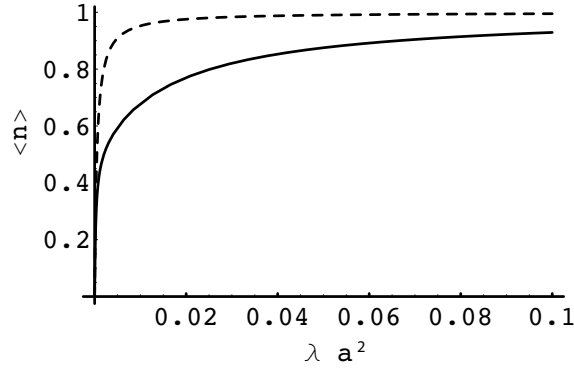


Figure 4. The fraction of occupied sites as a function of λa^2 for two lines separated by a distance $d = 0.1a$ with $\rho ad/2 = 1$ (solid). This is compared to the fraction of occupied sites for a single line of adsorption sites at the same background density (dashed line).

4. Average density

In this section, we will compute the average counterion density at an arbitrary point \mathbf{R}_0 . We can find the density directly by fixing the position of one of the ions. This will require the computation of the quantity

$$T_s(\mathbf{R}_0) = \rho^{s+1} \sum_{\mathbf{R}_1} \cdots \sum_{\mathbf{R}_s} \prod_{j=0}^s \rho_0(\mathbf{R}_j, -\mathbf{R}_{\sigma(j)}) \rho_X(\mathbf{R}_j, \mathbf{R}_{\sigma(j)}), \quad (26)$$

where the sum is over all s -cycles σ . Then the average density is given by [8]

$$\langle \rho(x_0, y_0) \rangle - \rho = \sum_{s=1}^{\infty} \frac{\lambda^s}{s!} T_s(\mathbf{R}_0), \quad (27)$$

where x_0 and y_0 are the components of \mathbf{R}_0 .

For $\pi \rho ad = 2\pi n$, n an integer, and for $\mathbf{R}_j = an_j \hat{x} + d\delta_j \hat{y}$, equation (26) decomposes into the product

$$T_s = \rho^{s+1} s! (-1)^s T_s^{(1)} T_s^{(2)}, \quad (28)$$

where

$$T_s^{(1)} = \sum_{n_1 \cdots n_s} e^{-\pi \rho x_0^2 / 2} e^{-(\pi \rho a^2 / 2) [n_1^2 + (n_1 - n_2)^2 + \cdots + (n_{s-1} - n_s)^2 + n_s^2]} e^{-\pi \rho a x_0 (n_1 + n_s) + i \pi \rho a y_0 (n_1 - n_s)} \quad (29)$$

and

$$T_s^{(2)} = \sum_{\delta_1 \cdots \delta_s} e^{-\pi \rho y_0^2 / 2} e^{-(\pi \rho d^2 / 2) [\delta_1^2 + (\delta_1 - \delta_2)^2 + \cdots + \delta_s^2]} e^{-\pi \rho d y_0 (\delta_1 + \delta_s)} e^{i \pi \rho d (\delta_1 - \delta_s)}. \quad (30)$$

Defining $z = x_0 + iy_0$ and \bar{z} its complex conjugate, we find that

$$T_s^{(1)} = \sum_{n_1 \cdots n_s} e^{-\pi \rho x_0^2/2} e^{-t[n_1^2 + \cdots + n_s^2 + (n_1 + \cdots + n_s)^2]} e^{-\pi \rho a(n_1 z - n_s \bar{z})}. \quad (31)$$

Using the transfer matrix \mathcal{T} and defining a new diagonal matrix,

$$\mathcal{M} = \begin{pmatrix} 1 & 0 \\ 0 & e^{-\pi \rho d^2/2 + \pi \rho d y_0 + i \pi \rho d x_0} \end{pmatrix}, \quad (32)$$

and its complex conjugate $\bar{\mathcal{M}}$, we find

$$T_s^{(2)} = e^{-\pi \rho y_0^2/2} \text{tr}(\mathcal{T}^{s-1} \bar{\mathcal{M}} \mathcal{T} \mathcal{M}) \quad (33)$$

for $s \geq 1$ and $T_0^{(2)} = e^{-\pi \rho y_0^2/2}$. Evaluating $T_s^{(2)}$ gives

$$T_s^{(2)} = [\lambda_+^{s-1} G_+(z) + \lambda_-^{s-1} G_-(z)], \quad (34)$$

where

$$4G_+(z) = \exp(-\pi \rho y_0^2/2) (\lambda_+ |1 + A|^2 + \lambda_- |1 - A|^2)$$

and

$$4G_-(z) = \exp(-\pi \rho y_0^2/2) (\lambda_- |1 + A|^2 + \lambda_+ |1 - A|^2),$$

and

$$A = \exp(-\pi \rho d^2/2 + \pi \rho d y_0 + i \pi \rho d x_0).$$

Using the results of Rosinberg et al. [8], we can compute $T_s^{(2)}$ in terms of

$$F(\zeta, z, t) = \frac{1}{2} \left\{ \theta_3 \left[\zeta + \frac{i \bar{z} t}{\pi a}, t \right] \theta_3 \left[\zeta + \frac{i z t}{\pi a}, t \right] + \theta_3 \left[\zeta - \frac{i \bar{z} t}{\pi a}, t \right] \theta_3 \left[\zeta - \frac{i z t}{\pi a}, t \right] \right\}.$$

This gives

$$T_s = (-1)^s s! \rho^{s+1} [\lambda_+^{s-1} G_+(z) + \lambda_-^{s-1} G_-(z)] e^{-\pi x_0^2/2} \int_0^1 d\zeta \theta_3^{s-1}(\zeta, t) F(\zeta, z, t). \quad (35)$$

Using equation (27), we find the average density

$$\begin{aligned} \langle \rho(z) \rangle - \rho &= -\lambda G_+(z) \rho^2 e^{-\pi \rho x_0^2} \int_0^1 d\zeta \frac{F(\zeta, z, t)}{1 + \lambda \rho \lambda_+ \theta_3(\zeta, t)} \\ &\quad - \lambda G_-(z) \rho^2 e^{-\pi \rho x_0^2} \int_0^1 d\zeta \frac{F(\zeta, z, t)}{1 + \lambda \rho \lambda_- \theta_3(\zeta, t)}. \end{aligned} \quad (36)$$

Finally, we note that

$$\begin{aligned} 4G_+(x_0, y_0) &= \lambda_+ \left\{ e^{-\pi \rho y_0^2} + e^{-\pi \rho (y_0 - d)^2} + 2e^{-\pi \rho [y_0^2 + (y_0 - d)^2]/2} \cos(\pi \rho d x_0) \right\} \\ &\quad + \lambda_- \left\{ e^{-\pi \rho y_0^2} + e^{-\pi \rho (y_0 - d)^2} - 2e^{-\pi \rho [y_0^2 + (y_0 - d)^2]/2} \cos(\pi \rho d x_0) \right\}, \end{aligned} \quad (37)$$

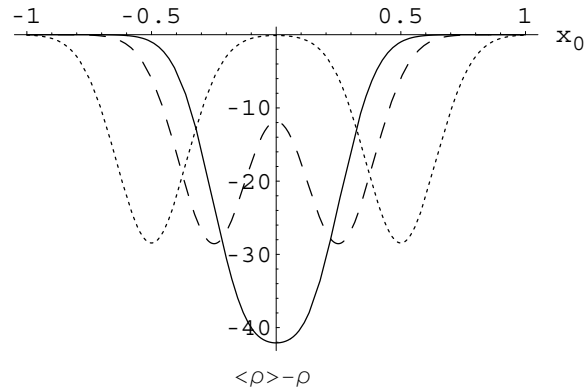


Figure 5. $\langle \rho \rangle - \rho$ as a function of y_0 for $x_0 = 0$ in units of a . Here $\rho ad = 2n$ and $\lambda\rho = 8$. We show $d = 0.25a$ (solid), $d = 0.5a$ (dashed), and $d = 1a$ (dotted).

and

$$4G_{-}(x_0, y_0) = \lambda_{-} \left\{ e^{-\pi\rho y_0^2} + e^{-\pi\rho(y_0-d)^2} + 2e^{-\pi\rho[y_0^2+(y_0-d)^2]/2} \cos(\pi\rho dx_0) \right\} \\ + \lambda_{+} \left\{ e^{-\pi\rho y_0^2} + e^{-\pi\rho(y_0-d)^2} - 2e^{-\pi\rho[y_0^2+(y_0-d)^2]/2} \cos(\pi\rho dx_0) \right\}. \quad (38)$$

Notice that, for $\pi\rho ad = 2\pi n$, the periodicity of $G_{\pm}(x_0, y_0)$ in x_0 is always commensurate with the lattice spacing a . In figure 5, we plot $\langle \rho \rangle - \rho$ as a function of y_0 for $x_0 = 0$, $\rho ad = 2n$, and $\lambda\rho = 8$ and for a variety of different spacings. Notice that the density has two peaks for large separations but a single peak as the separation d becomes smaller than the periodicity a . As a function of x_0 , the density is always periodic with period a .

5. Discussion

The main conclusion of our calculation is that for the geometry that we have chosen no attractive forces are induced by the discrete structure of the charged line.

The derivation of ΔF is valid for d taking values that are integer multiples of $2/(a\rho)$. Further, equation (21) gives the correct free energy for $d = 0$ (as we have already seen). It is conceivable, then, that equation (21) is correct for all values of ρ and d .

One of the features of this model that makes an exact evaluation possible is that a density ρ can be found such that the Ising variables δ_i and the integer variables n_i are uncoupled. When $\rho ad \neq 2m$ for an integer m , a coupling does indeed arise that makes the computation of the free energy more difficult. Additionally, if the adsorption sites are not aligned, an additional $\hat{x} \delta_i \Delta a$ component arises in \mathbf{R}_i , where Δa measures the degree of misalignment. This component introduces a coupling between the n_i and δ_i that will be discussed in future work.

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Взаємодія між двома рядами локалізованих адсорбційних центрів у двовимірній однокомпонентній плазмі.

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Ми розраховуємо вільну енергію для двох рядів локалізованих адсорбційних центрів, вставлених у двовимірну однокомпонентну плазму з густиною нейтралізуючого фону ρ . Енергія взаємодії між адсорбційними центрами є відштовхувальною. Ми також розраховуємо середнє число заповнення адсорбційних центрів і порівнюємо його з результатом для одного ряду центрів. Точний результат показує, що дискретизація не спричиняє зарядової асиметрії і не виникають сили притягання.

Ключові слова: однокомпонентна плазма, локалізована адсорбція

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