Correlation energies of simple bounded Coulomb lattices

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The lattice structure of bounded Coulomb systems is explored using a simple zero-temperature slab model. For slabs above a certain size infinite-volume behavior (i.e., a bcc lattice) occurs. Below this size surface effects dominate and the lattice type depends on the size, usually taking on an fcc-like symmetry. This state is similar to the lattice observed in recent computer simulations. These results have implications for experiments now underway.

In recent experiments,[1] a collection of N ions (where $N\sim 10^2-10^4$) are trapped and cooled to very low temperatures. The density n_0 and temperature T of this cloud of ions are such that the correlation parameter $\Gamma = e^2/a_{\rm WS}kT$ is much larger than unity, i.e., the ions are strongly correlated (here $a_{\rm WS}$ is the Wigner-Seitz radius given by $\frac{4}{3}\pi a_{\rm WS}^3 n_0 = 1$).

The regime of strong correlation has been studied extensively for an infinite homogeneous collection of ions. For instance, computer simulations[2] of such systems predict a phase transition to a body-centered-cubic (bcc) crystal at $\Gamma\simeq 178$. However, in the experiments the number of ions is sufficiently small that surface effects become important. In order to study these effects, a series of computer simulations modeling the trapped ions were performed.[3] The simulations show that the correlation properties are strongly affected by the boundedness of the cloud: at sufficiently large Γ the ions form concentric spheroidal shells whose symmetry is determined by the symmetry of the confining quadratic trap potential. Such shells have been seen in the experiments. On each shell, the simulations predict an imperfect two-dimensional (2D) hexagonal lattice. However, one would expect that for N sufficiently large a bcc lattice would form, perhaps in the center of the cloud, far from the boundaries.

This paper considers in more detail how the boundedness of the ion system affects the lattice structure, and an estimate is obtained for the minimum system size required before infinite-volume behavior is achieved. Several other relations are also derived, including the scaling of the number of shells as a function of the cloud size and shape, and the approximate spacing between shells.

In order to make theoretical progress, we focus here on the structure of the minimum energy ($T=0$) equilibrium state, and we consider a model which neglects the effects of the shell curvature but still incorporates the effects of boundedness. The model consists of a collection of ions trapped in planar geometry in a 1D quadratic well of the form $m\omega_z^2 z^2/2$ where m is the ion mass and ω_z is the oscillation frequency in the well. This system is therefore infinite and homogeneous in the x-y plane but bounded in the z direction. This model allows us to make predictions concerning the spacing and lattice structure of shells in large clouds where shell curvature is small compared to $a_{\rm WS}$ (However, shell curvature is an important element of the actual ion systems, so this model should be regarded as a simple first approximation).

Symmetry implies that the equilibria consist of a series of 2D lattice planes oriented parallel to the x-y plane. The number of planes P is determined for large P to be proportional to the total number of ions per unit x-y area, $\bar{\sigma}$. Sufficiently far from the surface planes, the lattice planes are evenly spaced, setting up a 3D lattice in the bulk. As $\bar{\sigma}\to\infty$, $P\to\infty$ so the system becomes homogeneous in z and, as is well known, the lattice with minimum energy has bcc symmetry. However, for finite P, surface effects are important; ion-ion correlations in the z direction are disrupted by the finite system size, and bcc symmetry is not necessarily minimum energy.

The effect of the surface on the structure of the $T=0$ state can be understood by considering the potential energy per ion of the system of stacked 2D lattice planes. The energy $e\Phi$ per ion may be written as $e\Phi = e\Phi_V + E_{\rm corr}$ where $e\Phi_V$ is the "Vlasov" energy per ion of a uniform slab of charge in the quadratic well and $E_{\rm corr}$ is the correlation energy per ion. The Vlasov energy is

$$e\Phi_V = [L\sigma/(4a_0) - \sigma^2/24]4\pi e^2/a_0 ,$$

where $\sigma = \bar{\sigma}a_0^2$ and $a_0 \equiv (4\pi e^2/m\omega_z^2)^{1/3}$. (In order to keep Φ_V finite we have assumed that the system is centered between conductors at $z=\pm L$.) Note that if correlations are neglected, a uniform density slab is an equilibrium since the quadratic well has the same confining effect as a uniform neutralizing background charge of density $n_0 = a_0^{-3}$. The ions therefore match their density to this background charge out to a value of z, set by σ, where the supply of ions is exhausted. The correlation energy $E_{\rm corr}$ is the additional energy due to the fact that the system is not uniform but is made up of separate lattice planes.

An analysis of the potential energy for a system of P 2D lattice planes yields the following expression for $E_{\rm corr}$:

$$E_{\rm corr}=\left[\frac{\sigma^2}{24P^2}+\frac{\sigma}{4P^2}\sum_{i=1}^{P}\sum_{\substack{j=1\\ j\neq i}}^{P}e^{-k|Z_i-Z_j|}\frac{\cos\mathbf{k}\cdot(\mathbf{X}_i-\mathbf{X}_j)}{ka_0}+\frac{1}{2P}\sum_{i=1}^{P}\left[\frac{\delta Z_i}{a_0}\right]^2\right]\frac{4\pi e^2}{a_0}+\frac{U_M}{2}\ , \tag{1}$$

where

$$U_M=\lim_{r\to 0}\left[\frac{2\pi e^2}{A_{\rm cell}}\sum_{\mathbf{k}\,(\neq 0)}\frac{\cos\mathbf{k}\cdot\mathbf{r}}{k}-\frac{e^2}{r}\right]$$

is the 2D Madelung energy associated with the interaction of an ion with its 2D lattice images. The sum over $\mathbf{k}$ is a sum over the reciprocal-lattice vectors for the 2D lattice, given in terms of the 2D primitive lattice vectors $\mathbf{a}$ and $\mathbf{b}$ by

$$\mathbf{k}=2\pi\hat{\mathbf{z}}\times(n\mathbf{a}+m\mathbf{b})/A_{\rm cell}$$

where $A_{\rm cell}=|\mathbf{a}\times\mathbf{b}|$ is the area of the primitive cell. The positions $(\mathbf{X}_1,\ldots,\mathbf{X}_p)$ are the positions of basis ions from which all other ion positions may be obtained through translations of the form $n\mathbf{a}+m\mathbf{b}$. Thus each value of $\mathbf{X}_i=(X_i,Y_i,Z_i)$ determines a 2D lattice plane through Z_i. In order to simplify the analysis we have focused on equilibria for which the 2D lattice symmetry is the same in each plane. The positions Z_i of the lattice planes may be found by consideration of electrostatic force balance; we find that $Z_i=Z_i^{(0)}+\delta Z_i$ where

$$Z_i^{(0)}/a_0=-[(P+1)/2+i]a_0^2/A_{\rm cell}$$

gives evenly spaced planes a distance $D\equiv a_0^3/A_{\rm cell}=\sigma a_0/P$ apart, and where δZ_i is given by

$$\delta Z_i=\frac{D}{2}\sum_{\substack{i,j\\ i\neq j}}\sum_{\mathbf{k}\,(\neq 0)}\mathrm{sgn}(Z_i-Z_j)e^{-k|Z_i-Z_j|}\times\cos\mathbf{k}\cdot(\mathbf{X}_i-\mathbf{X}_j)\ . \tag{2}$$

The terms in Eq. (1) have simple physical interpretations. The first term represents the difference between the Vlasov energy of a uniform density slab and that of a series of P sheets of uniform charge evenly spaced by distances D. The third term gives the correction due to nonuniformity of the spacing between the planes; Eq. (2) shows that this nonuniformity is a surface effect due to correlations between ions on different lattice planes ($\delta Z\to 0$ in the bulk). The last term is due to correlations between ions in a given plane, while the second term is due to correlations between different planes.

It is instructive to rearrange Eq. (1), writing

$$E_{\rm corr}=E_b+2E_s/P\ , \tag{3}$$

where E_b is the bulk energy per ion in the infinite lattice, obtained by setting $\delta Z_i=0$:

$$E_b=\left[\frac{D^2}{24}+\frac{D}{2}\sum_{i=1}^{\infty}\sum_{\mathbf{k}\,(\neq 0)}\frac{e^{-kDi}}{k}\cos\mathbf{k}\cdot(\mathbf{X}_{i+1}-\mathbf{X}_1)\right]\frac{4\pi e^2}{a_0^3}+\frac{U_M}{2}\ , \tag{4}$$

and the remaining energy gives the contribution due to the surfaces. The "surface energy" E_s is positive for stable lattices[4] and is independent of P for given D and sufficiently large P; the factor of 2 arises because there are two surfaces in the system.

The bcc lattice has the lowest value of E_b ($E_{\rm bcc}=-0.895\,929\,3e^2/a_{\rm WS}$) and so for $P\to\infty$ this is the equilibrium symmetry. However, other lattices, such as the face-centered-cubic (fcc) and hexagonal-close-packed (hcp) lattices, have values of E_b which are very close to the bcc value, so only slight differences in E_s are required in order to change the minimum energy symmetry. We will therefore be interested in equilibria which lead to these symmetric lattices in the bulk.

To determine the equilibria it is first instructive to make a simple approximation, first put forward by Totsuji and Barrat for cylindrical systems, which gives some insight into the form of the minimum energy solutions.[5] The approximation involves keeping the correlation energy U_M of an ion with its 2D lattice, and neglecting the other sum over $\mathbf{k}$ which represents interplane ion-ion correlations. The planes are then spaced evenly with $Z_i=Z_i^{(0)}$, and

$$E_{\rm corr}=\pi\sigma^2e^2/(6P^2a_0)+U_M/2\ .$$

Now, in general, U_M can be written as $U_M=-\alpha e^2/A_{\rm cell}^{1/2}$ where α is some dimensionless constant depending only on the lattice type (i.e., the angle between $\mathbf{a}$ and $\mathbf{b}$ and the ratio of their lengths).

Since $\sigma/P=a_0^2/A_{\rm cell}$ we can extremize $E_{\rm corr}$ with respect to P:

$$\left.\frac{\partial E_{\rm corr}}{\partial P}\right|_{\sigma,\alpha}=0=\left[\frac{-\pi\sigma^2}{3P^3}+\frac{\alpha\sigma^{1/2}}{4P^{3/2}}\right]\frac{e^2}{a_0}\ .$$

Thus the value of P which minimizes $E_{\rm corr}$ will be the integer closest to the minimum at $P_{\rm min}=(4\pi/3\alpha)^{2/3}\sigma$. The minimum possible value for $E_{\rm corr}$ is then, in this approximation, given by

$$E_{\rm corr}=-3(3\alpha^2/4\pi)^{2/3}e^2/(8a_{\rm WS})\ ,$$

where the Wigner-Seitz radius $a_{\rm WS}$ is related to a_0 by $a_{\rm WS}/a_0=(3/4\pi)^{1/3}$.

Now, it is well-known that the lattice with the maximum α value is the 2D hexagonal lattice, with $\alpha=3.921$.[5] Thus, by neglecting the interplane ion-ion correlations we find that the minimum energy configuration is a series of P evenly spaced 2D hexagonal lattice planes, where P is approximately $P_{\rm min}=1.045\sigma$

and $D/a_0=\sigma/P_{\min}=0.956$.

Note that interplane ion-ion correlations are disrupted in the ion clouds studied in the experiments because there are only a few shells, and also finite shell curvature impedes formation of a "commensurate" lattice from shell to shell. It is not surprising, therefore, that a 2D hexagonal lattice is seen in the shells (however, a more realistic model including finite T and curvature must be employed to fully explain the simulations). Furthermore, for relatively large clouds ($N \gtrsim 10^2$) the curvature of the shells is small compared to the intershell spacing, so the planar model for the shells is a useful approximation (although it does break down near the cloud center). The model predicts the spacing between shells to be approximately constant, given by D. This is observed in simulations and experiments. Such clouds are approximately ellipsoidal, with density $n_0=a_0^{-3}$ so $N=4\pi R^2 Z/(3a_0^3)$ where R and Z are radial and axial extents of the cloud, respectively. The approximate number of shells S as a function of N is thus

$$S \simeq (3NR/4\pi Z)^{1/3} a_0/D$$

for prolate clouds and

$$S \simeq (3NZ^2/4\pi R^2)^{1/3} a_0/D$$

for oblate clouds.

Unfortunately, the simple model put forward above fails in an important way. It is well known that as $P \to \infty$ the equilibrium structure is a bcc lattice. There is no transition to a bcc lattice in the above model because interplane ion-ion correlations have been neglected. In order to improve the model we now attempt to take these correlations into account.

Rather than attempt a full solution of this complex problem, we minimize E_{corr} for a few interesting cases. We consider the equilibria obtained from values of **a**, **b**, and (X_i, Y_i) given in Table I, as a function of D. [Symmetry implies that these values of (X_i, Y_i) are equilibrium positions.] These configurations are chosen because for certain values of D they correspond to bcc, fcc, or hcp lattices in the bulk, which all have low values of E_b. The bcc lattices are of particular interest since bcc symmetry is the correct result as $P \to \infty$. Lattices constructed from 2D hexagonal planes (entries 5, 6, and 7 of Table I) are also of interest since for $P=1$ this 2D lattice is the equilibrium symmetry.

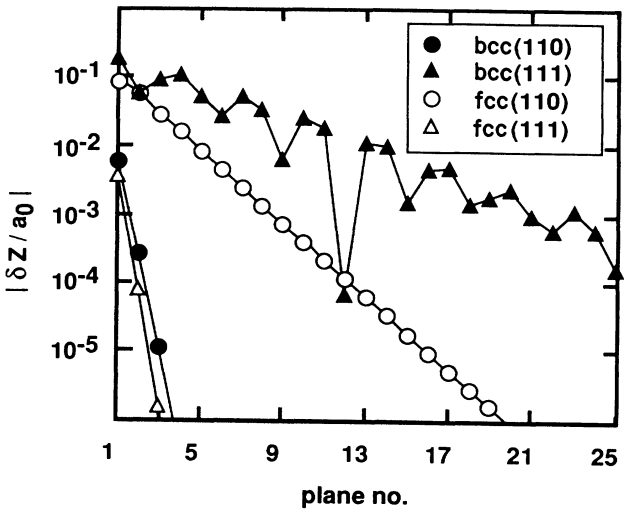

FIG. 1. Absolute value of δZ_i for various simple lattices. Plane 1 is the surface, and $P \gg 1$ is assumed, so the other surface does not interfere. (Lines connect points as an aid to the eye.)

The correlation energy for these lattices is found using Eq. (1), for given **a**, **b**, (X_i, Y_i), P, and D/a_0. The distance between lattice planes can be found by solving Eq. (2) for δZ_i, which is easily solved iteratively; solutions are displayed in Fig. 1 for some cases. Sufficiently far from the surface, $\delta Z \to 0$ so a 3D lattice is set up in the bulk. A plot of E_{corr} as a function of D/a_0 for various values of P is shown in Fig. 2 for the case of lattices given by entries 5 and 6. A general feature of the curves is that for large P minima occur at various values of D (labeled $D \equiv D_\infty$) corresponding to particularly symmetric bulk lattices. For instance, the minima in Fig. 2, at $D_\infty/a_0=2^{2/3}/\sqrt{3}$ and $D_\infty/a_0=2^{-2/3}/\sqrt{3}$, correspond to bulk fcc and bcc lattices, respectively, in which the (111) lattice planes are oriented parallel to the surface [referred to here as fcc (111) and bcc (111) lattices]. Values of E_b and E_s at D_∞ points are given in Table I. Figure 2 shows that as $P \to \infty$ the bcc (111) lattice has lowest energy (as expected), but even for quite large values of P, the fcc (111) lattice dominates. This is because the fcc (111) lattice has much smaller surface energy than the bcc

TABLE I. Correlation energies of bounded Coulomb lattices.

a	**b** (arb. units)	(X_i, Y_i)	D_∞/a_0	Lattice type	$E_b - E_{\text{bcc}}(e^2/a_{\text{WS}})$	$E_s(e^2/a_{\text{WS}})$
(1,0)	(0,1)	$(\frac{1}{2},\frac{1}{2})i$	0.6300	bcc (001)	0	2.3395×10^{-2}
(1,0)	(0,1)	$(\frac{1}{2},\frac{1}{2})i$	0.7937	fcc (001)	5.56×10^{-5}	1.0185×10^{-2}
(1,0)	$(\frac{1}{2},1/\sqrt{2})$	$(\frac{1}{2},0)i$	0.8909	bcc (110)	0	3.779×10^{-3}
(1,0)	$(0,\sqrt{2})$	$(\frac{1}{2},1/\sqrt{2})i$	0.5612	fcc (110)	5.56×10^{-5}	3.0370×10^{-2}
(1,0)	$(\frac{1}{2},\sqrt{3}/2)$	$(0,1/\sqrt{3})i$	0.3637	bcc (111)	0	4.6055×10^{-2}
(1,0)	$(\frac{1}{2},\sqrt{3}/2)$	$(0,1/\sqrt{3})i$	0.9165	fcc (111)	5.56×10^{-5}	2.147×10^{-3}
(1,0)	$(\frac{1}{2},\sqrt{3}/2)$	(0,0), i even $(0,1/\sqrt{3})i$, i odd	0.9165	hcp	9.11×10^{-5}	2.112×10^{-3}

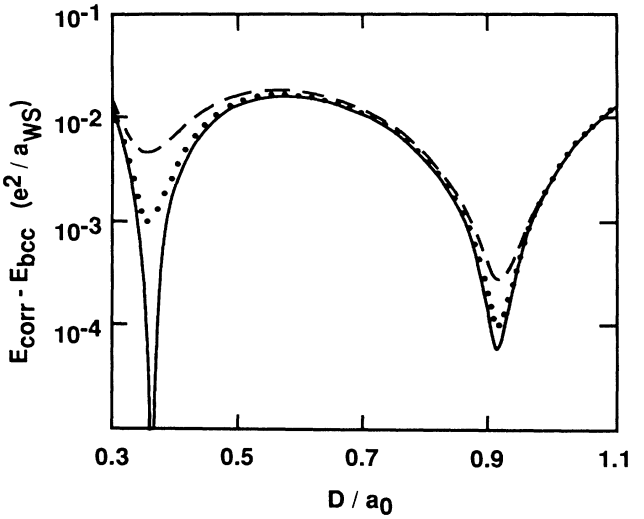

FIG. 2. E_{corr} vs D for the bounded lattice given by **a**, **b**, and (X_i, Y_i) of entries 5 and 6 in Table I. Dashes, $P=20$; dots, $P=100$; solid line, $P=\infty$.

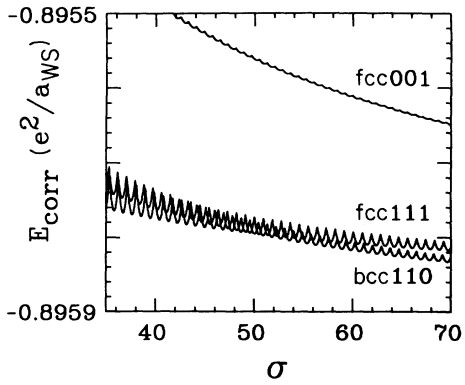

FIG. 3. E_{corr} vs σ for the bounded lattices given by **a**, **b**, and (X_i, Y_i) of entries 2, 3, and 6 of Table I. For each curve D varies around the D_∞ value of the lattice shown, and P varies by unity from cusp to cusp.

(111) lattice, since the lattice planes are spaced relatively far apart, reducing interplane correlations and localizing the effect of the surface to a few planes (see Fig. 1).

Note that for a given bulk lattice different lattice planes can be oriented parallel to the surface, giving rise to different surface energies. There are many such planes, labeled by various indices (l,m,n), but we consider only those planes with $l,m,n=0$ or 1. These are the most symmetrical cases and so are easiest to treat, and they also have the lowest surface energies since the planes are spaced relatively far apart.

The minimum energy state for a given value of σ is then found using Eq. (3). Consider two bounded lattices, 1 and 2, with different values of D_∞, $D_\infty^{(1)}$, and $D_\infty^{(2)}$, and with $E_b^{(1)} > E_b^{(2)}$. For each lattice minimum energy occurs for $P \simeq \sigma / D_\infty$. Equation (3) then predicts that lattice 1 has lower energy than lattice 2 for

$$\sigma \lesssim 2(E_s^{(2)} D_\infty^{(2)} - E_s^{(1)} D_\infty^{(1)})/(E_b^{(1)} - E_b^{(2)}) \ . \tag{5}$$

Now, E_b and E_s are functions of σ through the variable D, but for large σ, it is a useful approximation to take $D \sim D_\infty$ since $D = \sigma/P \simeq D_\infty$. In this case the values of E_s and E_b in Table I may be used in Eqs. (3) and (5), which then predict that for large σ the lowest energy bcc lattice is bcc (110), and the lowest energy fcc lattice is fcc (111). Furthermore, Eq. (5) predicts that this fcc lattice has lowest energy for $\sigma \lesssim 50$ (i.e., $P^{(bcc)} \lesssim 57$). An improved estimate for this transition is obtained by solving Eq. (3) exactly for E_{corr} for given σ and lattice type. This shows that the bcc-like symmetry corresponding to bcc (110) can in fact occur for $P^{(bcc)} < 57$ over certain narrow ranges of σ (see Fig. 3). However, for $P^{(bcc)} \gtrsim 60$ only this bcc-like structure occurs, in agreement with Eq. (5).

Thus, at $T=0$, surface contributions to the correlation energy affect the lattice structure for $\sigma \lesssim 50$ (or $P^{(bcc)} \lesssim 60$). Over most of this range a fcc-like lattice [corresponding to a fcc (111) lattice for particular values of σ] is minimum energy. This lattice consists of stacked 2D hexagonal planes and so is similar to the distorted lattice seen in simulations.[3] (The state is reminiscent of certain smectic mesophases observed in liquid crystals[6] which also consist of stacked 2D hexagonal planes, and in which interplane correlations are also either weak of nonexistent.) Of course, there may be other less symmetrical configurations, neglected in our simple approach, which lead to bulk bcc symmetry with lower surface energy. Furthermore, at finite temperature the entropy of the configurations is important, and curvature effects must be considered in order to make contact with the experiments. A more complete study of both slab and spheroidal equilibria is currently underway.

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