

Phase Transitions in Anisotropically Confined Ionic Crystals

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Crystalline, confined ionic systems exhibit well defined phase transitions as a function of the anisotropy of the confining potential. The transitions from one to two dimensions, from two to three, and back from three to two have been investigated as a function of this anisotropy with molecular dynamics simulations. The anisotropy at which such transitions occur seems to be proportional to a power of the number of confined ions.

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The simulation of cold, confined ionic systems, using the technique of molecular dynamics (MD) [1,2], has identified the type of ordering to be expected in such condensed systems, which now are beginning to be realized in storage devices [3-8]. These systems represent the rarest form of condensed matter (on the order of 10^8 ions/cm³) in nature. For larger systems [1] layered shell structure is expected and observed, while for smaller clusters of ions [2] unique shapes are obtained. In a cylindrical configuration, characteristic of storage rings [9], qualitative changes in structure are observed with increasing particle density; shape changes are also expected in the smaller systems with changing asymmetry in the confining potential.

The development of the technique of laser cooling in ion traps has led to the study of condensed crystalline [3-8] ionic systems in a number of trap configurations. In particular, for Paul traps (rf confinement) a "linear" configuration [7] has been used in which the confining field is strongly elongated, and a "ring" geometry [8] in which the plasma is confined to a toroidal volume of sufficient circumference to simulate the geometry of storage rings for fast particles. Progress has also been made in storage rings for fast heavy ions, with electron [10] and laser [11] cooling, but the temperatures required for crystallization have not yet been attained.

In the molecular dynamics simulation [1,12] of ions confined in a beam, distinct transitions in shape were found. With increasing linear particle density the configuration of cold ions forms first a string, then a two-dimensional zigzag pattern, and then a three-dimensional helix; finally new structures appear on the axis, and so a series of concentric cylindrical shells evolves. Recently these transitions were confirmed experimentally in the ring trap of Ref. [8].

In the simulations of isotropic confinement (where the Hamiltonian is exactly the same as J. J. Thompson's classical model of an atom), few (< 13) ions arrange themselves in simple geometrical shapes, as nearly equidistant from the origin as is feasible; with thirteen particles a second shell starts to form, and for large numbers of particles again concentric shells are seen in the simulations

[2]. Such shell structure has been confirmed experimentally in both geometries [5,8].

For anisotropic confinement (a rather usual occurrence in traps), changes occur in the shapes of these structures. When the confining force along one axis is sharply reduced, the lowest-energy configuration forms a line along that axis. If, in the other extreme, this component of the force is much stronger than the others, the ions will be confined to the plane perpendicular to the strong component. The transition points for such shape changes for the simplest systems of three and four ions are trivial and were given in Ref. [2].

A partial motivation of the present work was the observation [7], in the "linear Paul trap" configuration, that the number of ions that may reside on the axis of the trap is limited, and that at a certain point the ions bulge out in a two-dimensional zigzag pattern, similar to the one that was seen in simulations for storage rings. The objective of the present investigation is to determine the point where such transitions occur, as a function of the number of ions and of the anisotropy of the confining potential, and to map out the systematics of these dimensional phase transitions, to the point where the minimum-energy configurations change from one to two dimensions or from two to three dimensions.

For the simple case with cylindrical symmetry, the confining potential may be characterized by the ratio of the strengths of the confining potentials in the radial and the axial (z) directions,

$$\alpha = \omega_z^2 / \omega_{xy}^2, \quad (1)$$

where ω_{xy} and ω_z are the frequencies for radial or axial oscillations of a single particle in the confining potential. (Since the time average of the trapping potentials for static ions is well approximated by a harmonic force, the anisotropy may be characterized in terms of these frequencies.)

The potential energy of the system of N charges may then be written (following [2]) as

$$U = q^2 \sum_{i=1}^N \sum_{j=i+1}^N \left[\frac{1}{r_{ij}} + \frac{1}{2d^3} [x_i^2 + y_i^2 + \alpha z_i^2] \right],$$

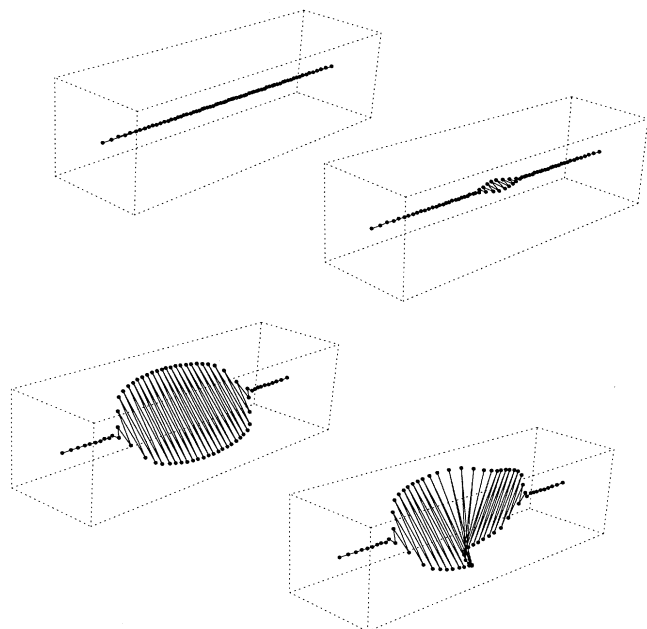


FIG. 1. Minimum-energy configurations of 70 ions with the asymmetry in the confining potential $\alpha = (1.52, 1.56, 2.17, \text{ and } 3.13) \times 10^{-3}$. The scales in the x and y directions are expanded by a factor of 100 with respect to that for the z axis.

where x_i, y_i, z_i are the ionic coordinates, $r_{ij} = [(x_i - x_j)^2 + (y_i - y_j)^2 + (z_i - z_j)^2]^{1/2}$ (d provides a unit of length, $d^3 = q^2/m\omega_{xy}^2$), and q and m are the ionic charge and mass. Since x, y, z , and r scale with d , this may be written as

$$U = \frac{q^2}{d} \sum_{i=1}^N \sum_{j=i+1}^N \left[\frac{1}{r'_{ij}} + [x_i'^2 + y_i'^2 + az_i'^2] \right], \quad (2)$$

where x', y', z' , and r' are the coordinates divided by d . The minimum-energy configurations therefore depend only on α and not on the other parameters.

Qualitatively, three transitions in the dimensionality of the configurations may be expected as a function of increasing α : (a) from a linear array along the axis to a two-dimensional zigzag pattern, still along the z axis; (b) from two-dimensional to three-dimensional configurations where the planar zigzag pattern begins to twist out of the plane; and (c) from a three-dimensional configuration back to a two-dimensional one, a roughly disk-shaped array in the equatorial x - y plane.

The transitions (a) and (b) are related to those seen in the infinite cylindrically symmetric system [1,12,13] at certain values of the linear particle density. But with the harmonic confinement in the z direction, the linear density of particles along this axis is not constant; it will also increase with increasing ion number.

The molecular dynamics program has been described previously [1]. Tests were made in order to ascertain the independence of the qualitative features of the results

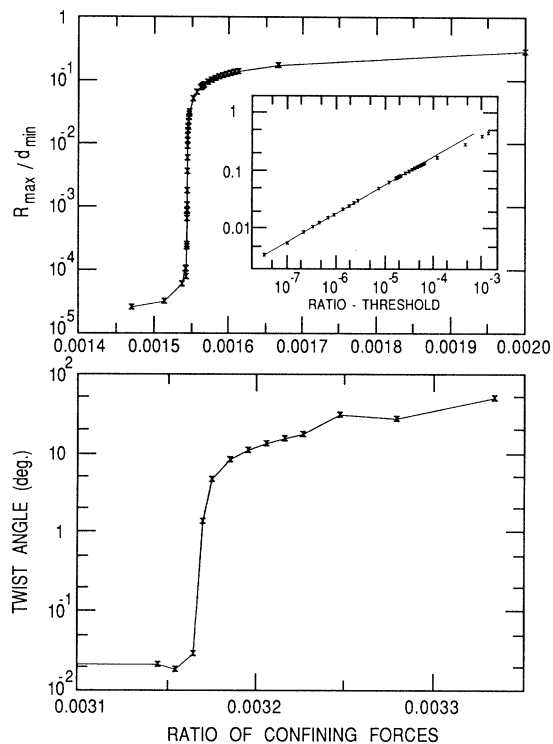


FIG. 2. The maximum radius $(x^2 + y^2)^{1/2}$ (divided by the least spacing along the z axis) as a function of the asymmetry α in the confining potential is shown in the top panel. The inset shows the same quantity plotted logarithmically for the asymmetry above the apparent threshold value, and showing a square-root threshold behavior. The lower panel shows the angle by which the zigzag pattern shifts out of a planar configuration.

from initial conditions, the time steps, and the rate of cooling in the simulations. The calculations were carried out for transitions (a) and (b) for several values of α as a function of ion number, and for 10, 25, 70, 180, and 500 ions as a function of α . Some typical results are shown in Fig. 1 for 70 particles. Figure 2 shows the maximum radius of the 70-ion configuration as a function of α , and the "twist angle" or the total amount by which the zigzag pattern twists out of a planar configuration as was shown in Fig. 1. It is clear that these dimensional transitions are indeed well defined and may be characterized as phase transitions. The very small residual fluctuations are small thermal effects from finite temperatures and/or step sizes in the MD calculations.

Figure 3 shows the properties of 500 particles above and below the transition (c). These transitions were also investigated for 10, 25, 70, 180, and 500 particles as a function of the parameter α . An interesting additional property of transition (c) is also seen. For the higher value, the system is strictly two dimensional and the inner region arranges itself in equilateral triangles; that is the characteristic of infinite two-dimensional Coulomb sys-

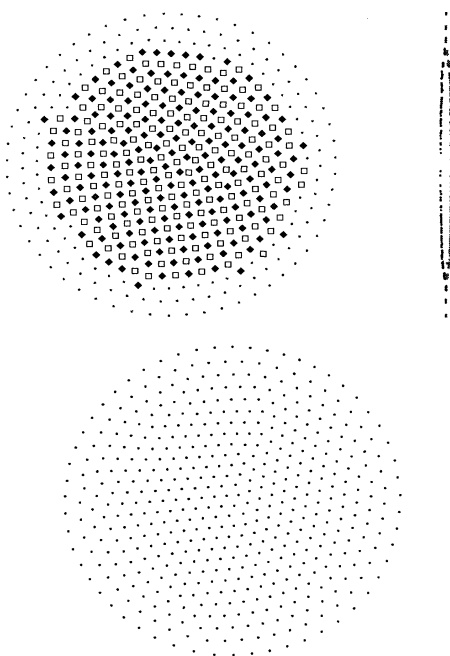


FIG. 3. The pattern of 500 particles for two values of α corresponding to above and below transition (c). For α of 50, shown in the lower part, all ions are in one plane. For α of 20, the ions in the central region form two layers as shown in the top part, with the symbols differentiating the ions in the layers. On the top right all the ions are plotted in projection showing their radius and z coordinates. Within the constraints of the boundary conditions, the ordering in the top part is in interlocking squares, and in the bottom part in equilateral triangles.

tems. However, when the ions separate into two layers [somewhat analogous to the zigzag pattern of transition (a)] the inner region seems to prefer a square pattern, with the two layers interlocking as in two layers of a bcc crystal. Thus the phase transition is also associated with the type of ordering seen in the x - y plane. This form of ordering, though, is transitory, because as the confinement approaches isotropy, the spherical (or near-spherical) shells show patterns with equilateral triangles rather than squares.

For the case where the confining forces are unequal along all three axes ($\omega_x \neq \omega_y$), the same result is obtained for transition (a) (from one to two dimensions), with the smaller of ω_x or ω_y determining the transition point, and the zigzag pattern oriented toward that direction. Transitions (b) and (c) have not been investigated for cases where the confining field is different along all three axes.

The dependence of the value of the asymmetry parameter α at which the transitions occur as a function of particle number is shown in Fig. 4. For the larger ($N > \sim 100$) systems, finding the true minimum-energy configuration is not easy in molecular dynamics near the transition point, and there is some uncertainty (perhaps a

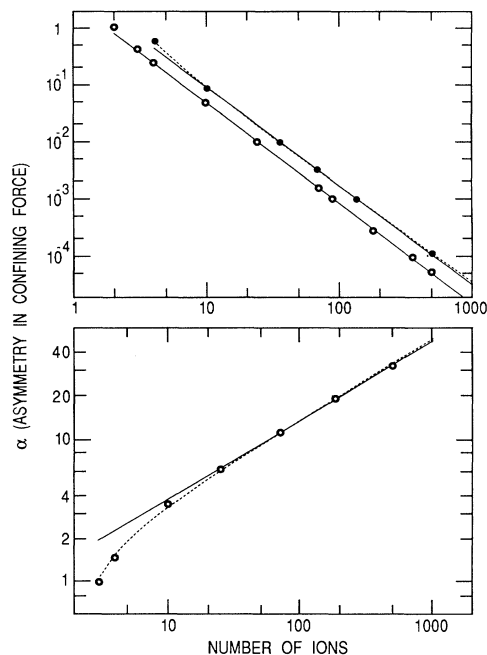


FIG. 4. Values of the transition points obtained from the MD simulations. In the top panel, the points for the transition (a) from one to two dimensions are plotted (open) and for the transition (b) from two to three (solid). In the bottom panel the points are for transitions (c) from three dimensions back to two. The lines represent the best-fit power-law behavior to this trend; the dashed lines represent the fit with a power of $(N-1)^\beta$ and $(N-2)^\beta$ for transitions (b) and (c).

few percent in α) for the values in the larger systems—no more than the size of the points in Fig. 4. Transitions similar to type (a) and (b) occur in the simulations of beams at particular values of the linear density λ (in terms of a unit of length defined by the plasma density) which have also been derived analytically [12]. Although the definition of λ is not quite appropriate here, one may use the radial focusing strength to obtain an equivalent value. The transitions seem to occur at the point when the linear density of the innermost pair is close to this value—but the changing linear density of the present systems does not permit a detailed comparison. No analytic form has been found for calculating the central spacing in a linear configuration, except for very small systems.

Included in Fig. 4 are values for the transitions for three and four particles which were obtained analytically in Ref. [2]. It seems clear that the general behavior is well described by a simple power law,

$$\alpha_{\text{tr}} = cN^\beta, \quad (3)$$

where α_{tr} is the value of the asymmetry parameter at which the phase transition takes place for N ions, and c and β are constants. The values describing the lines

shown in the figure are -1.73 , -1.76 , and 0.55 for β , and 0.395 , 0.178 , and 1.073 for c . The reason for such a simple relationship and the apparent similarity of the exponents for transitions (a) and (b) is not understood at present. A somewhat better fit is obtained for transition (c) if there is an offset to the number of ions and the relation $\alpha_{tr} = c(N-2)^\beta$ is used, as is shown in Fig. 4. Searching on such an offset also slightly improves the fit for transition (b), using $(N-1)^\beta$ but this is of doubtful significance. For transition (a) the best fit corresponds to no offset. The values of the exponent β with these offsets for transitions (b) and (c) are -1.68 and 0.52 .

To conclude, a systematic trend has been observed for the transitions changing the dimensionality in simple ionic configurations. The numerical behavior of these phase transitions remains to be explained. The presence of such transitions sets limits on the trap configurations that can produce certain classes of ordered ionic systems that may be of particular interest.

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