ANL/PHY/CP-95452

Ordering In Classical Coulombic Systems

JOHN P. SCHIFFER
Physics Division, Argonne National Laboratory, 9700 S. Cass Avenue, Argonne, IL 60439 and The University of Chicago, 5720 S. Ellis Avenue, Chicago, IL 60637

I would like to discuss the properties of classical Coulombic matter at low temperatures. It has been well known for some time [1,2] that infinite Coulombic matter will crystallize in body-centered cubic form when the quantity Γ (the dimensionless ratio of the average two-particle Coulomb energy to the kinetic energy per particle) is larger than ~175. But the systems of such particles that have been produced in the laboratory in ion traps, or ion beams, are *finite* with surfaces defined by the boundary conditions that have to be satisfied. This results in ion clouds with sharply defined curved surfaces, and interior structures that show up as a set of concentric layers that are parallel to the outer surface. The ordering does not appear to be cubic, but the charges on each shell exhibit a 'hexatic' pattern of equilateral triangles that is the characteristic of liquid crystals. The curvature of the surfaces prevents the structures on successive shells from interlocking in any simple fashion. This class of structures was first found in simulations [3] and later in experiments [4].

The Molecular Dynamics simulations are straightforward application of Newtonian mechanics with the equations of motion integrated in sufficiently small time steps to approximate the classical trajectories. At each step the interactions between all particles have to be computed anew, thus for an N-particle system $N^2/2$ terms contributing to the net force need to be computed. The method is limited by computer power, but modern parallel computers have been making considerable strides recently.

For small aggregations of ions isotropically confined in a harmonic potential the calculations are almost trivial, yet a number of the minimum-energy configurations have been reported incorrectly until recently [5,6]. This Hamiltonian corresponds to J. J. Thomson's classical pre-quantum-mechanics model of the atom and the configurations are shown in figure 1. Note that the charges are equidistant from the origin up to 12 ions, but the 13th ion prefers to sit at the origin. This has little to do with the symmetric icosahedron -- it is simply the consequence of 12 being the last integer smaller than 4π. Since the minimum energy configuration requires the ions to be as nearly equidistant as possible, the 13th ion sits at the center. For larger numbers of ions both the outer shell grows further, and slowly more ions join the first one in the interior until for 61 ions the last one again sits at the origin with two shells outside it.

With larger ion clouds the configurations are qualitatively similar: a set of concentric shells with approximate order on each one but no particular order between shells except for constant spacing. The case of 20000 ions in isotropic confinement is shown in figure 2 where 18 concentric shells are discernible in the system. Is there a sharp phase transition as a function of temperature for such systems? The answer is in figure 3 which shows the gradual cooling of a cloud of 1000 ions in isotropic confinement. It seems that an outer crust forms first at the surface of the space-charge limited ion cloud, then the interior layers form gradually, the shells becoming sharper as the temperature is lowered, but the shell widths seem to reach a limit, with the outer shell substantially sharper than the others. No sharp change in the shell structure is found with temperature in the simulations.

Now when the ion clouds become anisotropic the cloud takes on a spheroidal shape. But this is not just a matter of concentric spheroids as the consideration of simple

DISCLAIMER

systems will show. In the limit, for a relatively very weak restoring force along the z axis, all ions will sit on this axis along a line, though not equally spaced. As this force is increased, there is a distinct point where a two-dimensional configuration is favored, and the ions form a zig-zag pattern, starting at the center. With the force increasing further, another point causes a three-dimensional configuration to be favored. These sharp transitions show all the characteristics of *dimensional phase transitions* [7] and are illustrated for 70 ions in figure 4. In the limit of a relatively very strong force in z the system will again become a two-dimensional pancake at $z = 0$.

For large systems with anisotropic fields there are many more complicated transitions -- and a cloud can have linear tails, and go through the transitions from two to three-dimensional structures moving along its axis. A limiting case of interest is where the restoring force in one direction disappears, but a constant density of charges per unit length is maintained, such as in a beam of ions. In this case, the system is very similar -- with a set of concentric cylindrical shells [8].

How large a cloud is needed before the shells give way to the cubic structure characteristic of infinite systems? Recent experiments reported here with very large ($N > 100000$) ion clouds indicate conclusively that an appreciable fraction of the cloud is in a single body-centered cubic crystals. Simulations have not yet caught up with this, largely because for 100000 ions 10^{10} pairwise interactions must be computed at each time step and this is rather expensive of computer time. Short cuts, such as cutoffs or multipole approximations are dangerous -- since the competition between two symmetries depends on the fine details of the energies.

The simple normal modes of these ion clouds [9] are hydrodynamic multipole modes. One of them, the monopole mode or (for non-magnetic traps) volume oscillation, is a true eigenmode of these systems and proceeds without damping. Others (in non-magnetic confinement the volume-conserving shape oscillations), are illustrated in figure 5 and show damping, and this damping is a result of the mixing of the multipole modes with the true eigenmodes of the system. In other words, the multipole modes are those of a charged liquid -- the discrete structure of these clouds only enters into the damping. There are, however, also some (strongly damped) torsional modes that do depend on the discrete ordered structure, since a liquid would not support a shear displacement.

The question of temperature in these systems is an interesting one. In a rotating system, such as a Penning trap, the ion cloud is rapidly rotating and thus in a non-inertial system. The 'temperature' then seems to be the random component of motion in the rotating frame. In the radio-frequency Paul traps the ions are subject to an alternating rf field and the time average of this rf field gives a net confining Hamiltonian. The ions, however, undergo coherent oscillations in this rf field. The question of defining what is meant by a temperature -- from the perspective of the ordering phenomena, for instance, is clearly somewhat fuzzy -- especially since the coherent motion includes shearing movements in different directions between ions and their neighbors. Defining temperatures in these systems is rather delicate -- as a practical matter it is usually done by looking at the velocity spread in the direction that is not affected by the motion (e.g. along the magnetic field in a Penning trap, or perpendicular to the macroscopic motion in rf confinement.)

Quantum effects are not significant in these large clouds. If one were to cool these systems to μK regime it would reach its quantum-mechanical ground state. However the wave functions of the individual ions would extend over a volume that is very small compared to the inter-ionic spacing (typically tens of microns) and so this would be difficult to observe.

This research was supported by the U.S. Department of Energy, Nuclear Physics Division, under Contract W-31-109-Eng-38.

References

[1] S. G. Brush, H. L. Sahlin, and E. Teller, J. Chem. Phys. 45, 2102 (1966)
[2] E. L. Pollock and J. P. Hansen, Phys. Rev. A 8, 3110 (1973)
[3] A. Rahman and J. P. Schiffer, Phys. Rev. Lett. 57, 1133 (1986)
[4] S. L. Gilbert, J. J. Bollinger, and D. J. Wineland, Phys. Rev. Lett. 60, 2022 (1988)
[5] G. Birkl, S. Kassner, and H. Walther, Nature 357, 310 (1992)
[6] Robert Rafac, John P. Schiffer, Jeffrey S. Hangst, Daniel H. E. Dubin, and David J. Wales, Proc. Natl. Acad. Sci. 88, 483 (1991)
[7] J. P. Schiffer, Phy. Rev. Lett. 70, 818-821 (1993)
[8] R. W. Hasse and J. P. Schiffer, Annals of Physics 203, 441 (1990)
[9] Daniel H. E. Dubin and J. P. Schiffer, Phys. Rev. E 53, 5249-5267 (1996)

Figure Captions

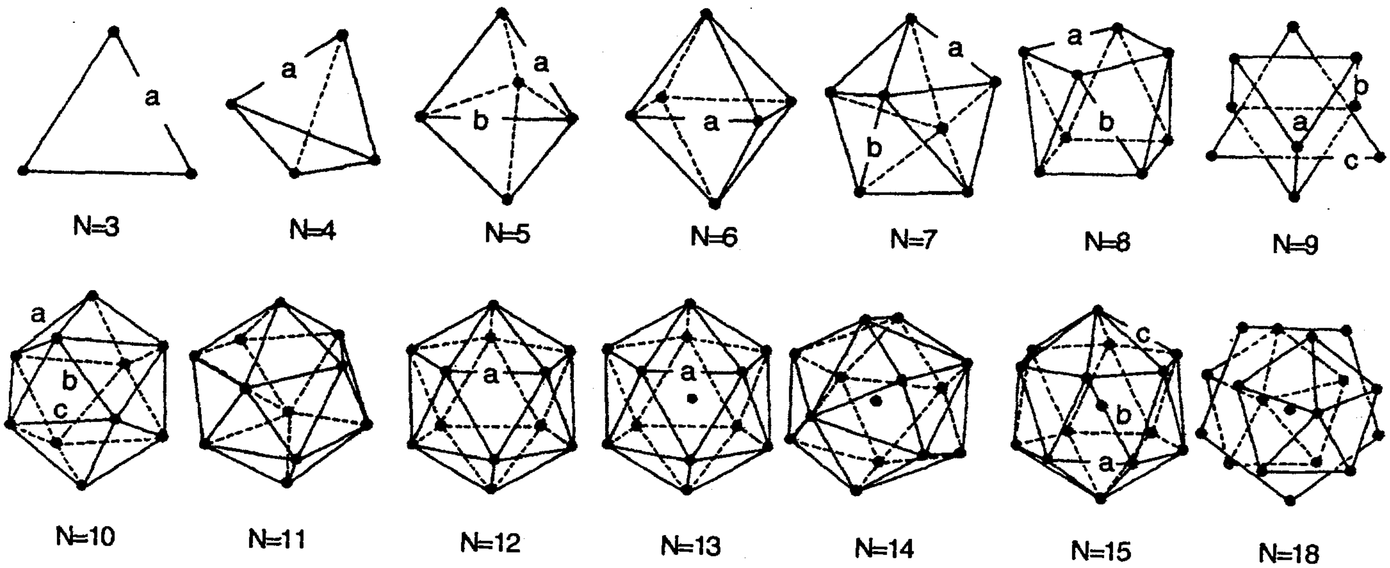
Fig. 1 Minimum energy configurations for few ions in isotropic confinement from ref. 6.

Fig. 2 View of outer surface, and interior radial density distribution of a simulation of 20000 ions in isotropic confinement.

Fig. 3 Radial density profiles of simulations of a 1000 ion cloud in isotropic confinement at various temperatures, corresponding approximately to $\Gamma = 0.1$, 10, 100, 10,000

Fig. 4 Simulation of the configurations of seventy ions in anisotropic confinement showing the dimensional transitions as the strength of the confining force in the longitudinal direction is increased, from ref. 7. The transverse dimensions are increased by a factor of ten compared to the longitudinal.

Fig. 5 Various hydrodynamic multipole oscillations induced in the simulation of a 1000 ion cloud. The top figure shows the decay in the amplitude of a monopole, quadupole and octupole oscillations. The bottom figure shows the decomposition of the quadrupole mode among the true eigenmodes of the system for a 100 and a 1000 ion cloud with the frequencies in units of the plasma frequency, from ref. 9.



a
N=3
a
N=4
a
b
N=5
a
N=6
a
b
N=7
a
b
N=8
b
a
c
N=9
a
b
c
N=10
N=11
a
N=12
a
N=13
N=14
c
b
a
N=15
N=18

Fig. 1

ANL-P-21,580

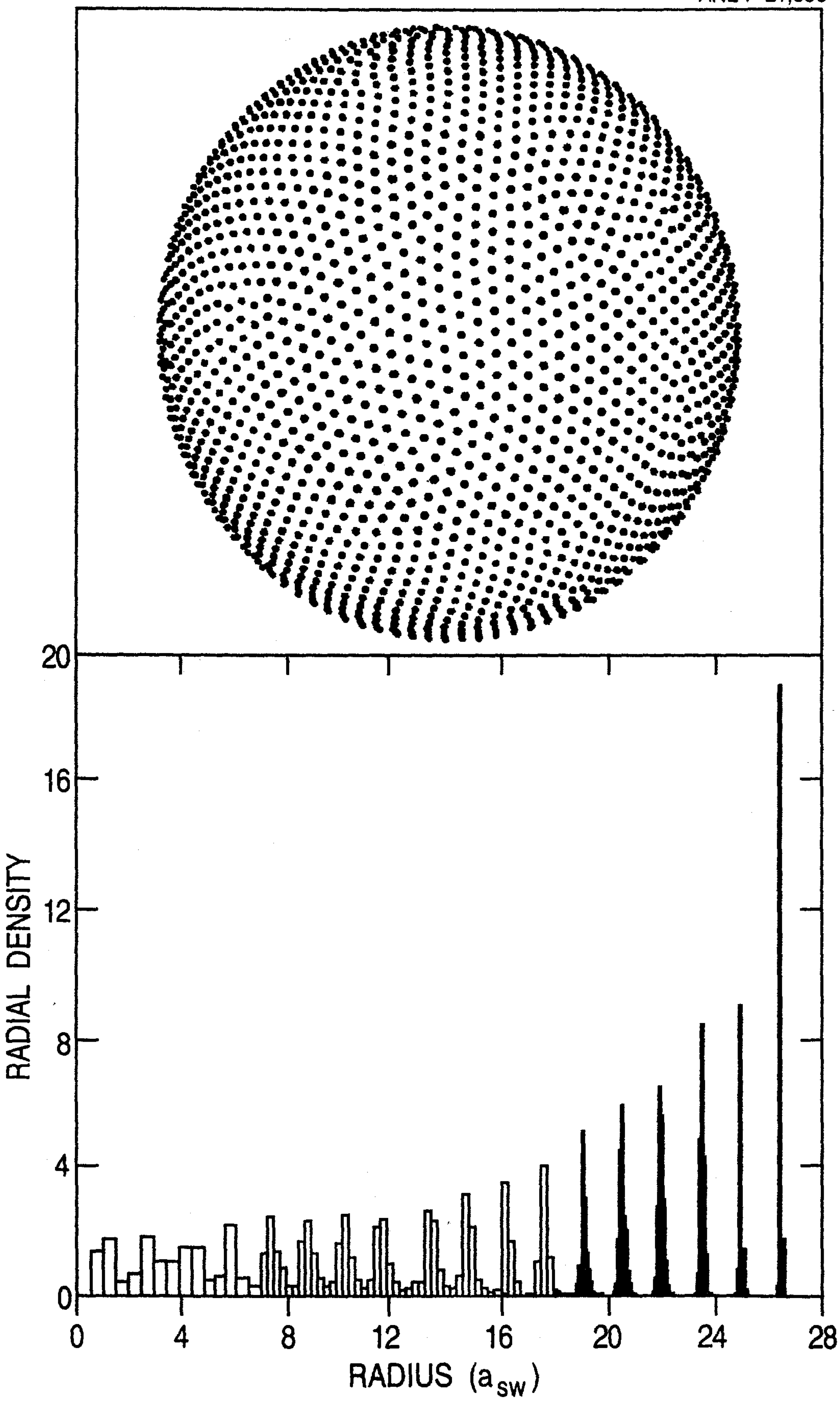


Fig. 2

ANL-P-22,249

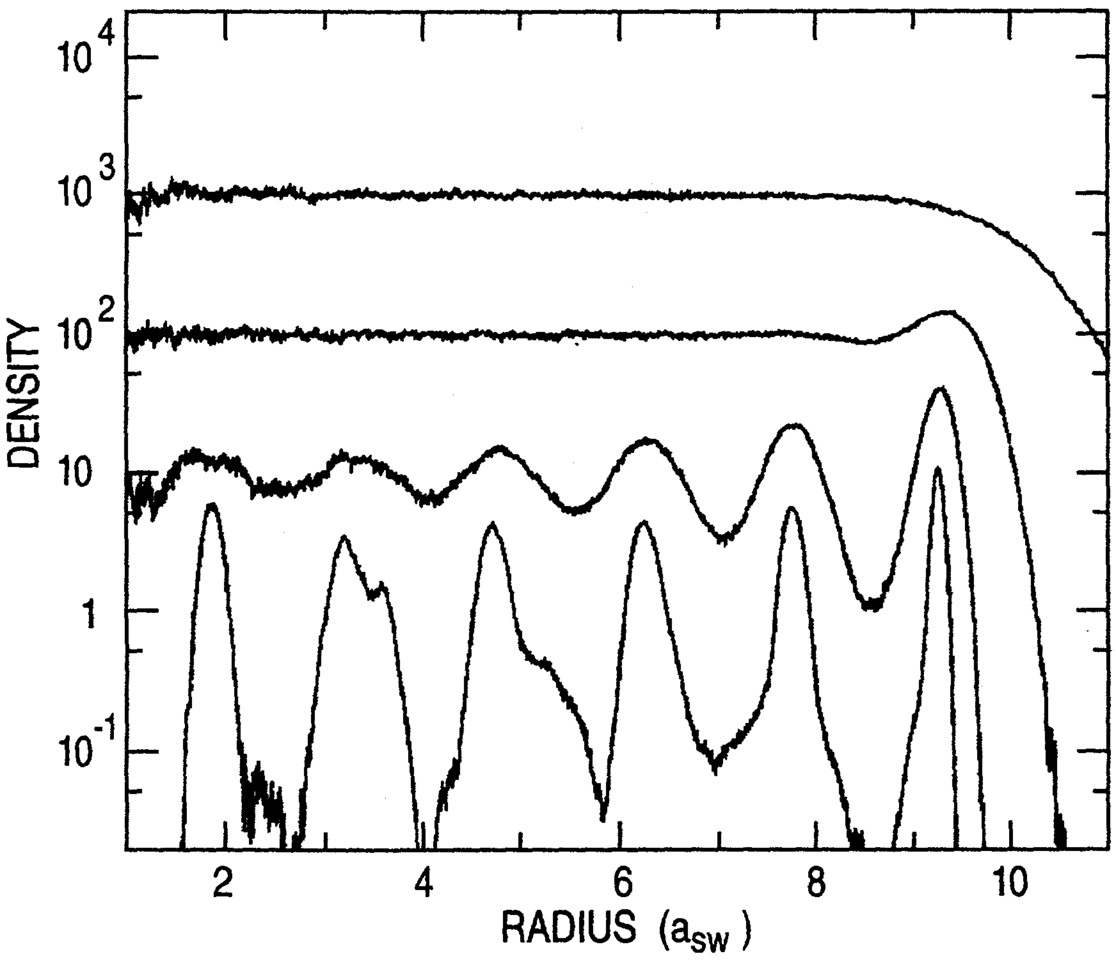


Fig. 3

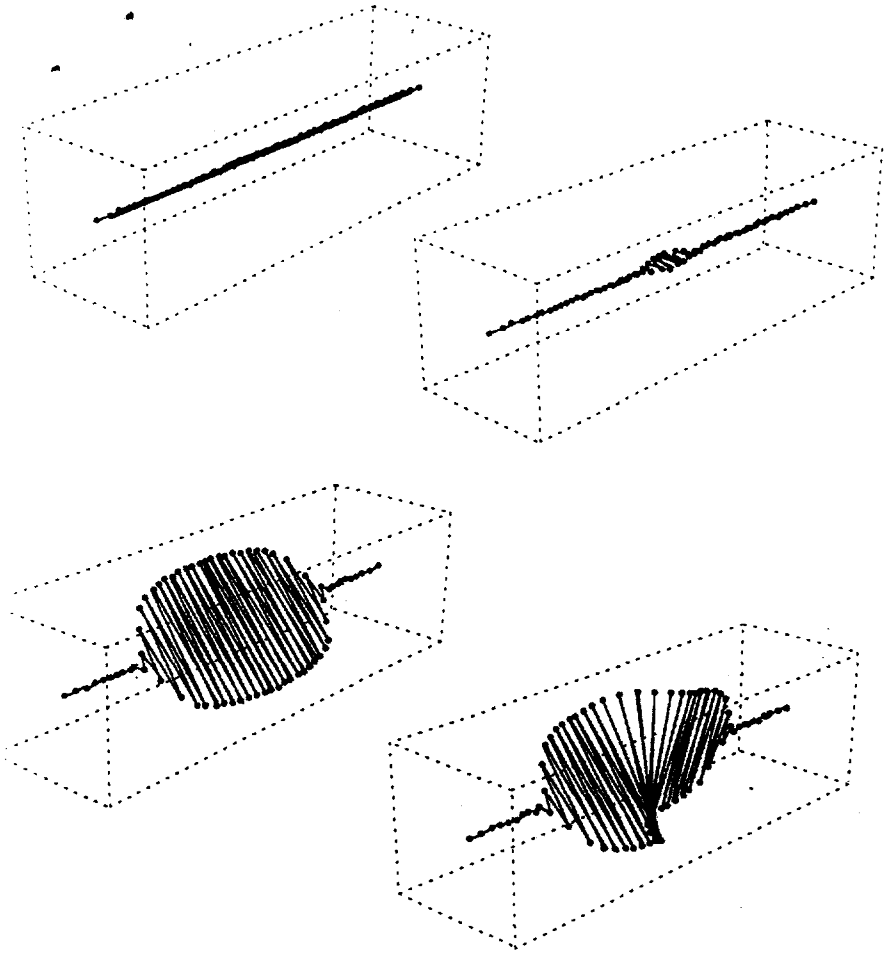
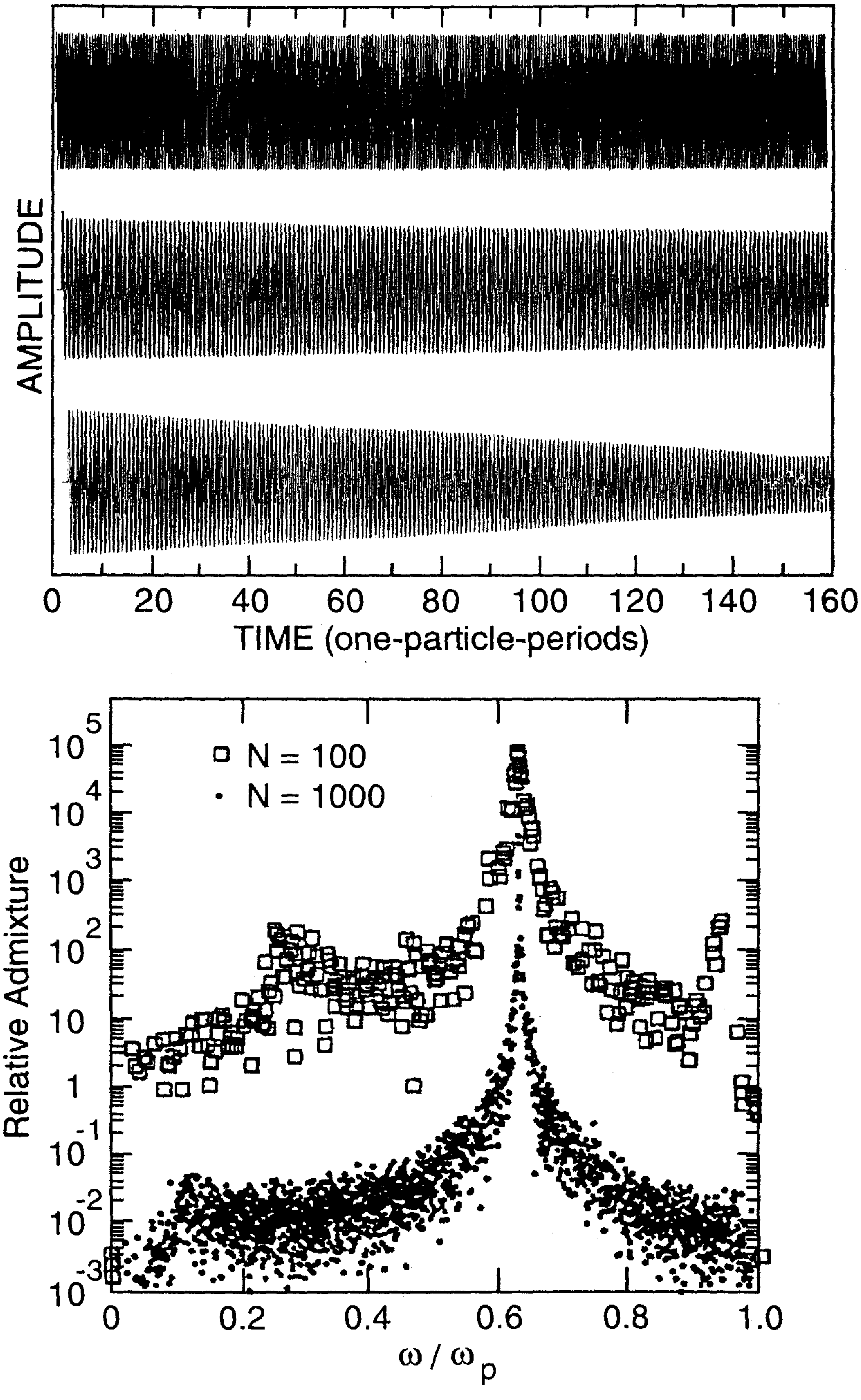


Fig. 4



AMPLITUDE
0
20
40
60
80
100
120
140
160
TIME (one-particle-periods)
Relative Admixture
10^5
10^4
10^3
10^2
10
1
10^{-1}
10^{-2}
10^{-3}
N = 100
N = 1000
0
0.2
0.4
0.6
0.8
1.0
ω / ω_p

Fig. 5