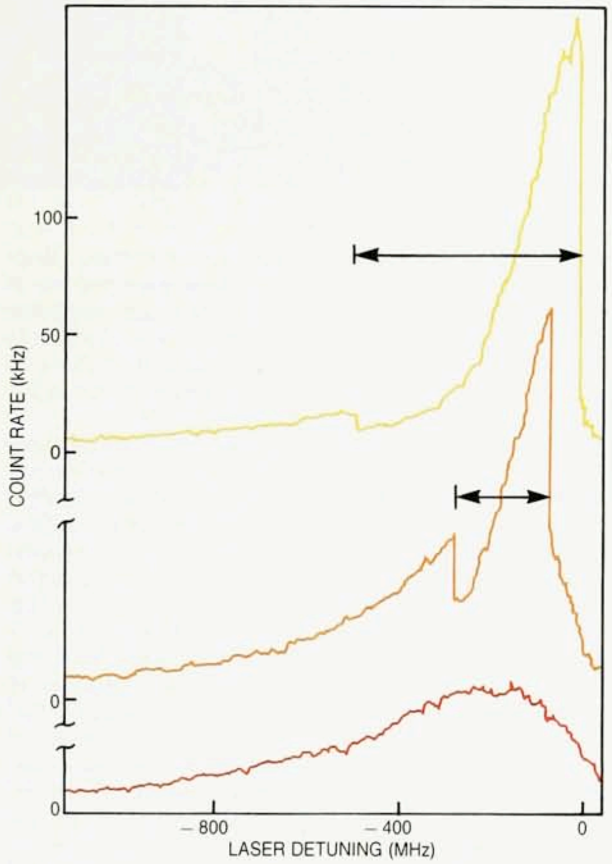
CLOUDS OF TRAPPED COOLED IONS CONDENSE INTO CRYSTALS

Photographs of ions trapped by electric and magnetic fields are revealing ordered structures ranging from a few ions in crystalline arrays to thousands of ions arranged on the surfaces of concentric shells. In some experiments with a handful of singly charged ions, varying certain parameters of the trap causes the regular structures to dissolve rapidly into clouds and just as rapidly to recrystallize—behavior resembling a phase change or an order-to-chaos transition. All these regular structures occur in systems with well-defined constituents and interactions. As such they can provide useful insights into collective phenomena such as cluster formation, Wigner crystallization and other strongly coupled plasmas.

Ordered arrays of trapped charged particles are not new. In 1959 Ralph F. Wuerker, Haywood Shelton (both at TRW, Redondo Beach, California) and Robert V. Langmuir (Caltech) photographed regular arrays of charged aluminum particles that were about 20 microns in diameter, and found that they successively melted and recrystallized. The recent experiments with ions, however, involve better-defined systems, whose particles have identical masses and charges. Furthermore, because individual ions have smaller charges than the aluminum particles, the new experiments can use far larger numbers of particles.



Excitation spectra from ions trapped by rf electric fields and cooled by laser radiation, as measured by the ion fluorescence. When the rf voltage is high (570 V), the ions move randomly in a cloud and the spectrum is broadened by the Doppler shifts (red curve). When the rf voltage is lowered below a certain threshold, the spectrum shifts suddenly to a sharp peak characteristic of an ordered state. In the orange and yellow curves, corresponding to rf voltages of 460 V and 360 V, respectively, arrows denote region of crystalline structure. The horizontal axis shows the amount by which the cooling laser is tuned below the resonant frequency. (Adapted from ref. 3.)

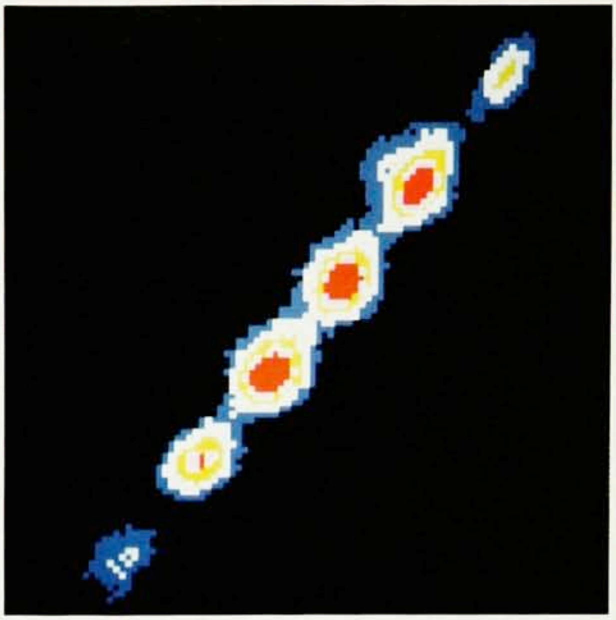
Cooling in a Paul trap

To study clusters with a small number of ions, the experimenters confine them with a configuration of electric fields known as a Paul trap. The effective confining potential results from applying a radiofrequency electric field between the end plates and a ring-shaped electrode in the center of the cylindrical trap. The electric quadrupole field has hyperbolic equipotentials in which the motion of ions is harmonic to first order.

The thermal motion of the ions is damped by laser cooling. (See the article by David J. Wineland and Wayne M. Itano in PHYSICS TODAY, June 1987, page 34.) In this technique, the ions are illuminated by a laser beam at a frequency just below one of the absorption lines of the ions. By the Doppler effect, the laser light appears at a higher frequency when the ions, jostling in thermal motion, move toward the beam direction. Because the radiation appears to these ions to be at resonance, the ions absorb the radiation and are slowed by conservation of momentum. Subsequent spontaneous emission of the radiation is isotropic and, on average, does not change the ions' momentum. Ions have been cooled below 10 mK by this technique. The ions can be imaged using the fluorescence produced as they re-emit the absorbed radiation. Laser cooling was proposed in 1975 by Theodor Hänsch (now at the University of Munich and the Max Planck Institute for Quantum Optics, Garching, West Germany) and Arthur Schawlow (Stanford University) and, independently, by Wineland (now at the National Bureau of Standards, Boulder, Colorado) and Hans Dehmelt (University of Washington).

The behavior of a system of trapped, cooled ions depends on the Coulomb coupling constant Γ, which is the ratio of the Coulomb interaction energy between neighboring, singly charged ions to their mean thermal energy. When Γ is greater than 1, the system enters the strong-coupling region. For very large values of Γ, a collection of ions is expected to arrange itself in a regular array. With the thermal motion so greatly reduced, each particle is essentially pinned at a point where the trap forces pulling it toward the center just balance the Coulomb repulsion forces from the other ions pushing it away from the center.

In 1980 Werner Neuhauser, Martin Hohenstatt and Peter Toschek of the University of Heidelberg, together with Dehmelt, photographed several ions in a Paul trap and found that the size of the two-ion image agreed with the expected equilibrium distance.[1] However, they did not have sufficient resolution to detect any possible structure.



Six mercury ions align along the z axis in a Paul trap for certain values of the trap voltage. Each ion is pinned at a point where the Coulomb forces pushing it outward are just balanced by the trap forces pulling it inward. In this false-color photograph ions are preferentially located in the red areas, which are the regions of most intense fluorescence. Neighboring ions are separated by several microns. (Courtesy of the National Bureau of Standards.)

From clouds to crystals

Last year, at the Max Planck Institute for Quantum Optics, a group led by Herbert Walther found evidence of ordered structures in systems containing 2 to 50 magnesium ions in a Paul trap.[2,3] Walther's fellow experimenters are Frank Diedrich, Ekkehard Peik, Jan Min Chen and Wolfgang Quint. They studied changes in the intensity of the fluorescence emitted by ions as they varied the laser detuning, that is, the amount by which the frequency of the cooling laser beam falls below the absorption line.

When a few ions move randomly in a cloud, the fluorescent spectrum is expected to be quite broad because of the Doppler shifts. The experimenters find such broad spectra when the rf field applied to the trap has a voltage high enough to cause the ions to have considerable thermal motion. (See the figure on page 17.) However, at a lower value of rf field intensity, the fluorescent spectrum jumps discontinuously to the sharply peaked shape associated with a single ion. This jump occurs as the magnitude of the laser detuning is decreased below a certain value. The Max Planck group interprets this behavior as a phase transition from a cloud-like state to a crystalline state in which motions are correlated. The narrow fluorescence peak in the crystal implies that the ions have assumed relatively fixed positions. At still smaller values of the laser detuning, a second jump in the spectrum suggests a transition back to the ion cloud.

The Max Planck group is able to induce transitions between clouds and crystals not only by varying the laser detuning but also by altering either the power of the cooling laser radiation or the magnitude of the rf voltage. In all of these cases, they find hysteresis effects expected for a phase transition: For example, jumps from the crystalline state to the cloud-like state always occur at higher rf voltages than transitions in the opposite direction.

Walther and his colleagues confirmed the transitions between states by observing the system visually. They imaged the fluorescence with a photon-counting system and videotaped the images. At the expected value of laser detuning, they saw the ion cloud suddenly crystallize, with individual ions clearly resolved. The transition occurs in the less than 0.04 sec between successive frames. The photograph on the cover shows one of the ordered structures they observed, a seven-ion configuration in which neighboring ions are about 20 microns apart.

Also last year, a group headed by Wineland at the National Bureau of Standards trapped and photographed a number of ions in regular crystalline arrays such as rings or linear configurations.[4] (See the photograph above.) The NBS team calls these structures "pseudomolecules." The separation distances between ions in these pseudomolecules are on the order of several microns—much greater than the spacings between atoms in a real molecule. The NBS group members include James Bergquist, Itano, John Bollinger and Charles Manney. In their experiments on Hg^+, they cool the ions to temperatures below 8 mK, corresponding to a Γ value of about 500. The crystalline structures seen at NBS agree with the configurations that would minimize the potential energy for a given ring voltage, according to calculations by Daniel Dubin and Thomas O'Neil (University of California, San Diego). In these calculations the configurations change abruptly with the ring voltage. Dubin points out, however, that the term "phase transition" is not precisely correct for a finite system at finite temperatures.

The NBS experimenters studied in some detail a pseudomolecule consisting of pairs of Hg^+ ions. They determined a particular absorption line for an individual Hg^+ ion and deduced the vibrational frequency of the pseudomolecule from the measured sidebands, which reflect the Doppler shift in the absorption frequency caused by the ion motion. The value of vibrational frequency determined in this way agreed well with theoretical predictions.

Toschek and his colleagues at the University of Hamburg—Th. Sauter, H. Gilhaus, Neuhauser and R. Blatt—have found metastable vibrational states of a single barium ion in a Paul trap,[5] and have also photographed clusters of two, three and four ions of that same element. They recently reported an observation of two novel cooling schemes for trapped particles.

Nature of the transitions

John Hoffnagle, Ralph DeVoe and Richard Brewer (all of IBM's Almaden Research Center in California) together with Luis Reyna (IBM T. J. Watson Research Center, Yorktown Heights, New York), have analyzed the behavior observed by Walther and his colleagues in terms of transitions from order to chaos.[6] The IBM group asserts that the relevant equations of motion contain ingredients that should lead to chaos: a radiofrequency driving term, dissipation due to laser cooling, and a Coulomb term that couples the ions nonlinearly. Brewer argues that the noise from fluctuations in ion recoil after spontaneous emission establishes the initial conditions that displace the ions from their equilibrium positions and start them on their path to chaos. He believes the noise term is small enough that it plays no role in the subsequent, deterministic development of chaos.

Brewer and his colleagues analyzed in two dimensions the simplest possible system—two barium ions in a Paul trap—and traced its evolution as a function of a parameter q, which depends, among other things, on the rf voltage. As q increases, the motions in the radial and axial directions become more strongly coupled until, at a critical value of q, the amplitudes of both motions suddenly increase and exhibit an erratic temporal and spatial dependence, characteristic of the chaotic state. The researchers find that the separation between two initially adjacent points in phase space diverges exponentially, as expected in chaotic systems.

The IBM team made direct measurements and photographed images of the behavior of two Ba^+ ions for comparison with their model predictions. They found that the transition from an ordered to a disordered state occurs in the real system at the q value predicted in their model. At a lower value of q the physical system recrystallizes; the model, however, does not exhibit condensation at this point because of a problem in truncation.

In related work, the IBM group has identified a heating source they feel is important. They find that Doppler shifts caused by the oscillation of ions at the frequency of the rf trap voltage can change the effect of the laser from cooling to heating. The main heating mechanism previously identified in Paul traps was rf heating.

The Max Planck investigators, joined by institute colleagues Reinhold Blümel and Wolfgang Schleich and by Yuen-Ron Shen of the University of California, Berkeley, have used three-dimensional molecular dynamics calculations to simulate the motion of any number of ions in a Paul trap.[7] From these simulations they can extract the excitation spectra and the jumps in them, and also reproduce the hysteresis loops seen in the fluorescence. Their calculations predict the value of the control parameter at which condensation from the cloud phase to the crystal phase occurs. The theoretical modeling does not indicate that an adiabatic change in the rf voltage will melt the crystals, although melting is observed in the experiments. However, the theory shows that the crystal may become very sensitive to fluctuations in the laser intensity, which could then trigger this transition.

This group explored the system dynamics and studied the relation between the stability of ion clouds and radiofrequency heating in Paul traps. The results suggest that the heating stems from deterministic chaos as ions in the chaotic cloud phase gain kinetic energy from the rf field that drives the ions in the Paul trap. The experimenters use this scenario to offer an explanation for the sharpness of the observed few-body phase transitions. The heating rate depends critically on the phase-space diameter of the ion configuration. When the ion separation is of the order of a typical ionic lattice constant, no heating occurs and the ions perform multiply periodic motion. Strong heating, however, sets in suddenly at a critical size of the ionic array. For very large clouds the heating rate becomes negligible, which again permits regular motion of the ions.

Pure ion plasmas

To study large collections of trapped ions, called pure ion plasmas, experimenters typically use a different type of trap. The Paul trap can hold only a certain number of ions before rf heating becomes too large for the ions to be successfully cooled. Studies of plasmas with several hundred to a few thousand ions cooled to temperatures below 10 mK have therefore been carried out with Penning traps, in which a static magnetic field, rather than the rf field of the Paul trap, helps to confine the ions.

A large number of ions stored in a trap is analogous to a one-component plasma, which consists of a single species of charges embedded in a uniform background charge of opposite sign. In a particle trap or storage ring, the trapping fields effectively play the role of the neutralizing background charge. A decade ago, O'Neil and John Malmberg of the University of California, San Diego, suggested that a pure electron plasma can be confined in a Penning trap and cooled to the cryogenic temperature range, where one expects to obtain liquid and crystal states. They showed that the thermal equilibrium of such a system is identical to that of a one-component plasma. With a simple sign change, these ideas also apply to a pure ion plasma.

For many years theoretical studies of one-component plasmas considered only systems of infinite extent, but the possibility of experimental studies on finite systems in the strong-coupling region has recently stimulated theorists to look at the effect of small size and realistic boundary conditions on the resulting structures. Rather than condensing into a body-centered cubic structure, as predicted for unbounded systems, a system with several hundred ions is expected to form concentric spheroidal shells. This general behavior was outlined in 1986 by the late Aneesur Rahman (University of Minnesota) and John P. Schiffer (Argonne National Lab and the University of Chicago) as part of molecular dynamics calculations they were doing to simulate the behavior of ions in storage rings.[8] In 1988 Dubin and O'Neil predicted this spheroidal structure specifically for the Penning trap.[9] The ions diffuse rather freely on the surfaces of these shells, but not between them. O'Neil explained to us that the system behaves like a liquid on the surfaces of the shells but like a solid between the shells. Such behavior resembles that of smectic liquid crystals. As temperatures are lowered further and the coupling gets stronger, the ions assume a hexagonal lattice structure on the shell surface.

Sarah Gilbert (NBS), Bollinger and Wineland have seen the predicted shell structure in experiments with clouds of beryllium ions in a Penning trap.[10] This team used a laser probe beam as well as two cooling beams, one perpendicular and one diagonal to the axis of the trap, which parallels the magnetic field. Each laser beam induces fluorescence and enables the experimenters to see sections of the shells that form. (See the photograph on page 20.) The structures they have seen range from a single shell with 20 ions to 16 shells with a total of 15 000 ions. The number of shells corresponds to what the theory predicts for the given number of ions. However, some of these shells had cylindrical rather than the predicted spheroidal shapes, a finding that is not yet understood.

Gilbert and her colleagues also explored the dynamical behavior of ions on the shells: They switched off the fluorescence from the ions in certain regions of the cloud by tuning the probe laser beam to a frequency corresponding to a nonfluorescing state. By tracing the diffusion of these "tagged" ions, they found that the ions within a shell moved distances of more than 100 microns in 0.1 sec, but that ions took several seconds to diffuse between shells.

Malmberg and his collaborators at San Diego have confined pure electron plasmas in Penning traps and cooled them to the cryogenic temperature range. These plasmas are much larger (10^{10} electrons) than the ion plasmas but the correlation strength achieved is lower. The San Diego group feels that the electron plasmas are probably large enough to exhibit the bulk properties of an unbounded one-component plasma, but that substantially higher Γ values must be reached before a crystal can be obtained.

Strings of beads

If ions can condense in electromagnetic traps, they might also crystallize in an ion storage ring, where the particles are similarly cooled and confined. In heavy-ion rings, where ionization levels are high, the Γ value could be orders of magnitude higher than it is for singly charged ions. Measurements made in 1980 at the NAP-M proton storage ring in Novosibirsk, USSR, hinted that the beam there had some coherent structure.[11] In their model of ions in a storage ring, Rahman and Schiffer found that when the density of beam particles is very high, the ions will arrange themselves on the surfaces of concentric cylinders, which wrap around the ring with the axis at their centers. On each cylindrical surface, the ions occupy points on a triangular grid. If the beam density is sufficiently low, the ions space themselves regularly along the axis, like a string of beads.

Everyone is now eager to look for these jewels in the heavy-ion storage rings now under construction at such places as GSI in Darmstadt, West Germany; the Max Planck Institute in Heidelberg, West Germany; and the University of Aarhus in Denmark. All are expected to produce beams below the temperature of 1 K that precluded further investigation of ion crystals at Novosibirsk. Dietrich Habs (Max Planck Institute for Nuclear Physics, Heidelberg) has discussed several models of the behavior of ions in these rings.[12] Schiffer told us that several features of real ion storage rings not included in the calculations he did with Rahman may make it more difficult to see the predicted structures. One is the time variation of the focusing fields. A more serious omission, he felt, was the curvature of the ring. (The model treats the ring as a straight cylinder.) Ions closer to and further from the ring's center have different travel times. This difference may introduce a shear and cause differential cooling across the beam. However, with appropriate cooling and some adjustments in parameters, the new ion rings may yet produce ordered beams.

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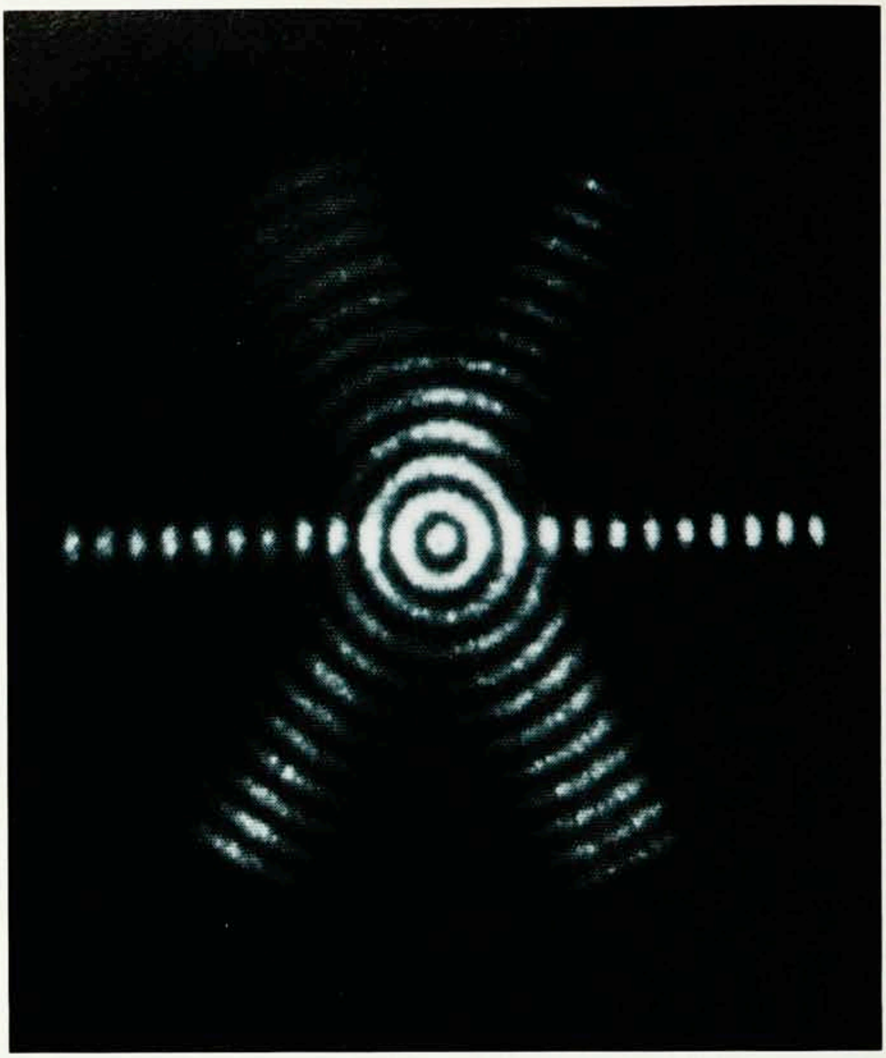

Arrangement in cylindrical shells of 15 000 ions in a Penning trap is evident in this photograph. The cross section of the shells is most visible where the laser beams (two for cooling and one for probing) cross the ion trap. The ions are found to arrange themselves on the surface of 11 concentric shells and a central column. Ions diffuse rather freely on the shell surfaces but not between shells. (From ref. 10.)

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