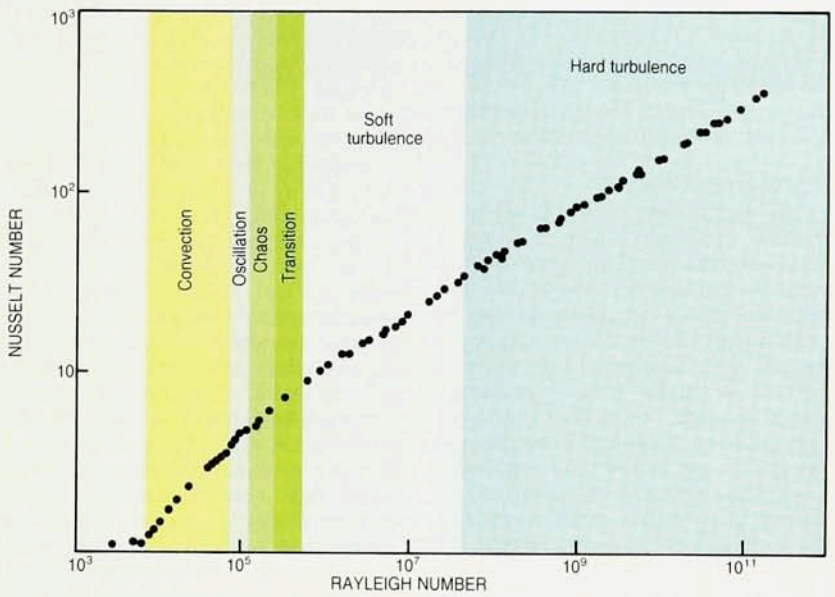
RAYLEIGH–BÉNARD EXPERIMENT PROBES TRANSITION FROM CHAOS TO TURBULENCE

Progress in understanding chaos—actually, temporal chaos—has been enhanced over the past ten years by the close agreement between some experiments and the theoretical descriptions of chaotic behavior. Our understanding of turbulence is not yet at the same level. However, a group led by Albert Libchaber at the University of Chicago has recently characterized with great accuracy turbulent flows in a Rayleigh–Bénard cell.[1] The Rayleigh–Bénard experiment studies convective flows in a fluid that is confined in a gravitational field between two parallel horizontal plates and heated from below. Earlier experiments on this system by Libchaber, among others, played an important role in our understanding of chaos.

The states of a classical system are represented as points in phase space. A classical system—for example, a fluid flow—is said to exhibit temporal chaos if two very similar states of the system become very different as the system evolves in time. More precisely, the distance between two points in phase space increases exponentially with time. Infinitesimal uncertainties in the knowledge of the initial state of a temporally chaotic system are therefore magnified exponentially, and it is impossible to determine the future state of the system exactly.

In a regular, or nonchaotic, system the dynamical variables are periodic or quasiperiodic functions of time. Therefore the power spectrum, which is the square of the Fourier transform in time, of a suitable dynamical variable shows sharp peaks at the natural frequencies. In a temporally chaotic system, by contrast, the power spectrum shows broad peaks above a noisy background that extends over a large frequency range.

Turbulent behavior is chaotic both in time and in space. Different parts of a system that exhibits temporal chaos behave coherently; in a turbulent system, on the other hand, the different regions show largely independent and incoherent chaotic behavior.



Nusselt number, the ratio of the total heat transported across a Rayleigh–Bénard cell to the heat transported by conduction, for Rayleigh numbers up to 10^{11}, as measured at the University of Chicago in a cell filled with helium. The curve has a slope of $1/3$ in the region marked "soft turbulence," but the slope decreases to a value of $2/7$ in the region marked "hard turbulence." Heat transport by convection therefore becomes less efficient in the hard-turbulence state.

Just as a chaotic system may be distinguished from a nonchaotic one by the power spectrum of a suitable dynamical variable, chaotic and turbulent systems may be distinguished from one another by the coherence function. The coherence function is a measure of the Fourier transform of the correlation in time between values of a dynamical variable at two different points in the fluid. Although it is impossible to predict in a temporally chaotic system the value of, say, the velocity at a point from knowledge of velocity values at that point up to some much earlier instant, values at the same instant but at different points in the system remain correlated in chaotic dynamics. The coherence function is therefore nonzero in a chaotic system. Even this so-called spatial coherence is partially lost in a turbulent system.

Temperature measurements at two different points in the Rayleigh–Bénard cell by the Chicago group—François Heslot, Bernard Castaing and Libchaber—in fact find two different turbulent states, which occur for different values of the Rayleigh number. (The Rayleigh number is a dimensionless parameter whose value determines the nature of fluid flows in the Rayleigh–Bénard cell.) In one of these states, which the Chicago group calls the soft-turbulence state, there is very little correlation between the temperature measurements at the two points, so the coherence function is zero. In the second, hard-turbulence state, the coherence function shows a peak at a certain frequency.

This indicates some oscillation in the system at that frequency. The two states also differ in the statistics of temperature fluctuations and in the efficiency with which heat is convected across the cell. "The emergence of a peak in the coherence function at large Rayleigh numbers is a most intriguing feature," Leo P. Kadanoff (University of Chicago) told us. Since the experiment, Kadanoff and theorists in his group at Chicago have collaborated with experimenters Xiao-Zhong Wu, Castaing, Heslot and Libchaber in extending ideas about the existence of boundary layers in turbulent systems and in developing a mathematical description of the hard-turbulence state. Besides Kadanoff, the Chicago theorists are Gemunu Gunaratne, Stefan Thomae, Stéphane Zaleski and Gianluigi Zanetti.[2]

Rayleigh number

Swirls and eddies in the flows in a channel, especially near an obstacle that obstructs the flow, figure prominently in the imagery used to describe turbulent flows. The channel may be a natural stream or a laboratory wind tunnel. But, Libchaber told us, it is difficult to make precise measurements in such "open flow" systems. Furthermore, turbulent flows depend sensitively on initial and boundary conditions, which are also difficult to control in open-flow experiments. In the Rayleigh–Bénard experiment, by contrast, the temperature difference between the top and bottom plates can be controlled with great precision. The simpler boundary conditions in the Rayleigh–Bénard experiment also make the theoretical analysis easier, at least near the onset of convection. For these reasons, the Rayleigh–Bénard experiment has emerged as a possible model system for studying chaotic and turbulent fluid flows. Libchaber added, however, that in the Rayleigh–Bénard cell it is difficult to reach the fully turbulent state, which is seen in open-flow systems.

Convective flow in the Rayleigh–Bénard cell is described by differential equations for the fluid velocity $\mathbf{v}(x,t)$, temperature $T(x,t)$ and pressure $P(x,t)$ at point x and time t. The differential equations follow when the conservation laws for mass, momentum and energy are applied to the motion of infinitesimal fluid elements. Solutions to the equations, and therefore the fluid flows, depend on the values of the Rayleigh number R, given by

$$R = \frac{\alpha g d \Delta}{D_T \nu}$$

and of another dimensionless parameter, the Prandtl number Pr, given by

$$\mathrm{Pr} = \frac{\nu}{D_T}$$

Here, α is the coefficient of thermal expansion; g is the acceleration due to gravity, d is the distance and Δ the temperature difference between the hot and cold plates of the Rayleigh–Bénard cell; D_T is the thermal diffusivity; and ν is the kinetic viscosity. Because both the thermal diffusivity and the kinetic viscosity vary inversely with the density, the Rayleigh number varies as the square of the fluid density. Rayleigh–Bénard experiments using different fluids but involving the same Rayleigh and Prandtl numbers show similar flows, because fluid parameters such as thermal conductivity and viscosity enter the equations only through those dimensionless numbers.

According to the analyses by Lord Rayleigh in 1916 and by S. Chandrasekhar in 1961, the conductive mode of heat transport between the lower (hotter) plate and the upper (cooler) plate of the Rayleigh–Bénard cell becomes unstable when the Rayleigh number exceeds a critical value, which is about 1708 for a horizontally infinite layer. The fluid in the cell will then transport heat between the plates not merely by conduction but also by convection. In convection, warm fluid rises up from the lower plate and then falls down as it loses energy due to viscous drag and diffusion of heat to cooler parts of the fluid. The upward and downward flows of the fluid constitute the Rayleigh–Bénard rolls. In conduction the fluid velocity is zero and the temperature decreases linearly with the distance from the lower plate; the fluid velocity and temperature in Rayleigh–Bénard rolls obey the hydrodynamic equations. The wavelength, or the characteristic distance between the upward and downward fluid flows, is on the order of twice the distance between the plates, and it depends weakly on the Rayleigh number.

A state of perfect convection rolls is not chaotic. It corresponds to a steady-state solution to the basic equations. When stable rolls in the Rayleigh–Bénard cell are convecting heat between the hotter and the cooler plates, temperature measurements at any point show no time dependence. But the rolls develop many kinds of instabilities as the Rayleigh number is increased. For example, under suitable conditions and for a range of Rayleigh numbers, the temperature at a point in the cell shows an oscillatory time dependence; the power spectrum then shows sharp peaks at frequencies characteristic of the oscillations. The number of distinct frequencies in the power spectrum increases, and the peak amplitudes decrease, as the Rayleigh number is successively raised above certain values. When the Rayleigh number is about an order of magnitude higher than the value at the onset of convection, the peaks disappear entirely and the power spectrum shows noise-like fluctuations over most of the frequency range. This is the so-called period-doubling route to chaos, whose universal properties Mitchell Feigenbaum (now at Rockfeller University) discovered. This route to chaos, which is one of many explored in the past 20 years, has been extensively studied in the Rayleigh–Bénard cell. (See PHYSICS TODAY, April 1986, page 17.)

The Chicago experiment

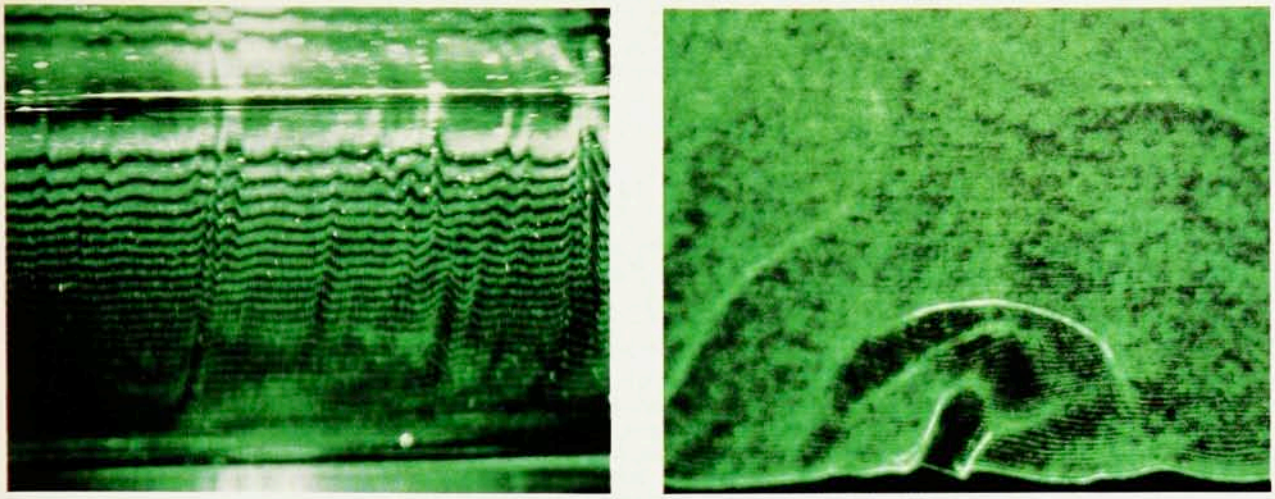
Several experiments during the last 50 years have studied the efficiency with which a fluid transports heat by convection. A measure of this efficiency is the Nusselt number, the ratio between the total heat flux and that due to conduction. Before the onset of convection, the Nusselt number is 1. As the Rayleigh number exceeds the critical value for the onset of convection, the Nusselt number becomes greater than 1; it then is usually represented as

$$N \approx (R - R_c)^{\beta}$$

where R_c is the Rayleigh number at the onset of convection. For Rayleigh numbers slightly greater than the critical value for the onset of convection, β is 1.

Experiments over the past 30 years have found values close to $\frac{1}{3}$ for β as the Rayleigh number was increased far beyond the value for the onset of convection, suggesting that the convective flows show a different scaling behavior at large Rayleigh numbers. The Chicago group studied both the Nusselt number and the time-dependent temperature fluctuations to characterize the different flows microscopically. The experiment spanned a range of Rayleigh numbers—from about 10 to about 10^{11}. The Prandtl number did not change significantly as the Rayleigh number was varied.

The Chicago group used helium gas above its critical point in a cylindrical cell whose diameter was equal to its height. The horizontal plates were made of thick copper; the vertical walls were thin and made of stainless steel. This choice of cell materials is fairly standard nowadays, and it ensures that heat is transported efficiently from the horizontal plates to



Waves and plumes on the boundary layer at the hot plate of a Rayleigh–Bénard cell filled with water. Left: A sheet of light composed of approximately 40 interference fringes was shone at grazing incidence on the bottom plate. The picture shows the pattern of light and dark strips reflected from the plate. The pattern is deformed from the one in the incident sheet because the waves on the boundary layer deform the density profile. Right: A laser beam was shone on the back wall of a Rayleigh–Bénard cell, toward the viewer and normal to the direction of convective flow. The picture shows a projection of the transmitted light on a screen. The white lines around the plume are an optical effect that arises because the fluid in the plume is hotter. (Courtesy of the University of Chicago.)

the fluid and that it is not conducted from the bottom plate to the top plate by the vertical walls. The use of helium gas in the Rayleigh–Bénard cell to probe a wide range of Rayleigh numbers was pioneered in 1975 by David Charles Threlfall (then at Cambridge University).

The Rayleigh–Bénard experiment was done for six values of the gas density. The smallest and the largest of the six density values differed by more than three orders of magnitude. (The corresponding smallest and largest pressure values were 0.3 torr and 625 torrs.) At each density, the temperature difference between the plates could be changed by almost three orders of magnitude, from about 3 mK to 1 K. Thus the Rayleigh number, which varies as the square of the density and depends linearly on the temperature difference between the plates, could be varied by almost ten orders of magnitude. The convective flows in the cell were monitored for this wide range in Rayleigh numbers by measuring temperature using two bolometers, each about 200 μm in size. One of these bolometers was placed about 200 μm above the bottom plate and midway between the cell axis and the cell walls; the second bolometer was placed midway between the two plates and directly above the first bolometer.

The figure on page 17 shows the Nusselt numbers measured for Rayleigh numbers up to 10^{11} in the Chicago experiment. For a range of Rayleigh numbers between 5×10^5 and 4×10^7, β is $\frac{1}{3}$, in agreement with earlier measurements. Beyond this range of Rayleigh numbers, however, the curve in the figure bends downward, giving a value of 0.282 ± 0.006 (close to $\frac{2}{7}$) for β. Some earlier experiments for Rayleigh numbers greater than 4×10^7 had shown values of β slightly less than $\frac{1}{3}$. Nevertheless, the value the Chicago experimenters report for Rayleigh numbers greater than 4×10^7 came as a surprise to many experts. It was generally believed that a value close to $\frac{1}{3}$ was the asymptotic value for β for arbitrarily large Rayleigh numbers. To distinguish the behavior between Rayleigh numbers 5×10^5 and 4×10^7 from that for higher Rayleigh numbers (up to the values studied), the Chicago group calls the former soft turbulence and the latter hard turbulence. Guenter Ahlers (University of California, Santa Barbara) thinks that the values of the Rayleigh numbers for the onsets of soft and hard turbulence may depend on the geometry of the Rayleigh–Bénard cell.

The time dependences of temperature recordings, as well as their statistics, are very different in the two types of turbulent behaviors. The figure on page 20 shows histograms of temperature recordings from the bolometer placed midway between the plates. In soft turbulence, the probability of recording a given temperature value in the middle of the cell decreases like a Gaussian about some mean value; in hard turbulence, the probability decreases exponentially on either side of the mean value. This statistical behavior of temperature fluctuations is valid everywhere in the central region of the Rayleigh–Bénard cell.

In hard turbulence, both the mean value of the temperature recording and the width of the histogram depend on the Rayleigh number. But the probability function has a universal form: When the temperature is scaled by a factor that is a measure of the scale for temperature fluctuations, histograms for different Rayleigh numbers fall on top of one another. The characteristic temperature scale Δ_c varies with the Rayleigh number as

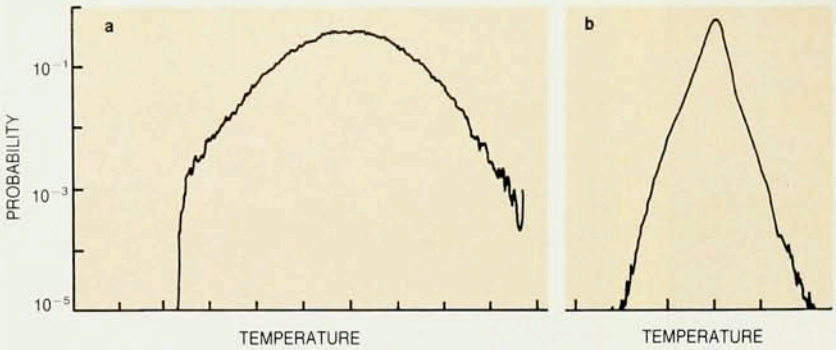
$$\frac{\Delta_c}{\Delta} \approx R^{\gamma}$$

The experiment finds a value of -0.147 ± 0.005, or about $-\frac{1}{7}$, for γ. The Chicago group also postulates that in hard turbulence the fluid in the middle of the cell has a characteristic velocity u_c. This velocity increases with the Rayleigh number according to

$$\frac{u_c d}{\nu} \approx R^{\epsilon}$$

An earlier, approximate experiment[3] measured a value of $\frac{3}{7}$ for ϵ.

As the distinction between chaos and turbulence would lead one to expect, the coherence function between the two bolometers begins to vanish in the transition region between chaos and soft turbulence. In the figure on page 17, this is the region in which the Rayleigh number



Histograms of temperature fluctuations show how often a temperature value is measured at a point in the middle of a Rayleigh–Bénard cell. The statistics, here shown on a semilog scale, are Gaussian for Rayleigh numbers between 5×10^5 and 4×10^7 (**a**), but change to an exponential form for higher Rayleigh numbers (**b**). Each unit on the temperature axis corresponds to $^1/_{20}$ of the temperature difference applied across the cell. The maxima in the histograms correspond to the mean temperature value in the cell's central region.

is between 2.5×10^5 and 5×10^5. The coherence function at all frequencies remains close to zero in soft turbulence. In hard turbulence, however, the coherence develops a peak at a characteristic frequency ω_p that scales with the Rayleigh number as

$$\frac{\omega_p d^2}{D_T}\approx R^{\delta}$$

The experiment finds a value of 0.491 ± 0.005 for δ.

The boundary layer

An explanation for the value of $^1/_3$ for β, the exponent that characterizes the dependence of the Nusselt number on the Rayleigh number, was proposed by C. H. B. Priestley and by Willem Malkus (MIT) in the 1950s. Malkus's ideas were further developed by Louis Howard (Florida State University, Tallhassee). According to these ideas, the applied temperature difference in the cell is distributed about equally between two thin layers of fluid, one at each plate. Ironically, heat is transported across the boundary layers by conduction, and although there is no temperature gradient in the cell's central region the fluid motion there is turbulent.

The heat flux due to conduction varies inversely with the distance between the points across which a temperature gradient is applied. When the temperature difference applied between the plates of the Rayleigh–Bénard cell a distance d apart is assumed to appear only across boundary layers of thickness λ, the Nusselt number must vary as

$$N\approx d/\lambda$$

The "$^1/_3$ law" for the Nusselt number therefore easily follows from dimensional analysis once one postulates the existence of the boundary layer, because the Rayleigh number varies as d^3.

The turbulent boundary layer

Kadanoff and his colleagues at Chicago have extended ideas about the existence of a boundary layer to determine exponents β, γ, δ and ϵ in hard turbulence. Some aspects of this work, especially those that establish relations between the exponents, are reminiscent of scaling ideas that Kadanoff, among others, proposed in the late 1960s to understand and derive the relations between exponents that characterize the thermodynamic singularities at a second-order phase transition.

According to the Chicago group, the boundary layer that forms near the plates in the Rayleigh–Bénard cell at large Rayleigh numbers is not a passive formation, just as the region in the cell's center is not entirely quiescent. Instead, the characteristic temperature Δ_c and speed u_c of fluid elements in the center of the cell must be such as to balance the heat flow across the boundary layer. Therefore the heat flow H across the cell is

$$H\approx\Delta_c u_c$$

which suggests that

$$\beta=\epsilon+\gamma$$

By dimensional arguments

$$u_c\approx(\alpha g d\Delta_c)^{1/2}$$

which gives, on using the definition of ϵ,

$$2\epsilon=\gamma+1$$

Thus only one of the three exponents β, γ and ϵ is independent. The Chicago group determines β by using the condition that the terminal velocity of a fluid element that breaks off from the boundary layer and rises to the central region of the cell must be the characteristic velocity u_c. The terminal velocity is obtained by balancing the viscous force against buoyancy. This balance gives

$$u_c\approx\frac{g\alpha\Delta\lambda^2}{\nu}$$

When combined with the scaling behavior of λ,

$$\lambda\approx R^{-\beta}d$$

this balance yields a second relation between β and ϵ. The two relations between β and ϵ are readily solved to give

$$\beta=^2/_7;\quad \epsilon=^3/_7$$

These values are in agreement with the experimental values.

The ideas about a turbulent boundary layer allow the Chicago group not only to derive values of the exponents, but to also estimate the values of the Rayleigh number for the onset of hard turbulence and for several other properties. The origin and significance of the peak in the coherence function in hard turbulence are not understood yet. But the theory predicts a value of $^1/_2$ for δ, the scaling exponent for the peak frequency, which is in good agreement with the experimental value.

The pictures on page 19 show the waves on the boundary layer and the plumes that emanate from it. These swirls and plumes, and their breaking off from the boundary layer and motion into the central region of the cell, are essential aspects of the theoretical ideas that the Chicago group has proposed to explain hard turbulence.

Victor Yakhot and Steven Orszag (Princeton University) have developed a renormalization-group procedure over the past few years to study turbulent flows. Yakhot and Orszag add a noise term in the hydrodynamic equations to account for the effect that flows on a large length scale have on those on shorter scales. The noise statistics can be modified to describe turbulent flows that show intermittency, that is, which have highly turbulent regions distributed sporadically in space.

In the theoretical scheme of Yakhot and Orszag, the Navier–Stokes equation with the noise term is approximately solved using renormalization-group techniques. Orszag told us that the method has been very successful in predicting basic constants of turbulent flows. Yakhot, in a recent calculation for the Rayleigh–Bénard cell,

reports values for the exponents, among other parameters, that also agree with those obtained experimentally.[4]

—ANIL KHURANA

References

1. F. Heslot, B. Castaing, A. Libchaber, Phys. Rev. A **36**, 5870 (1987).
2. B. Castaing, G. Gunaratne, F. Heslot, L. Kadanoff, A. Libchaber, S. Thomae, X.-Z. Wu, S. Zaleski, G. Zanetti, preprint.
3. H. Tanaka, H. Miyata, Int. J. Heat Mass Transfer **23**, 1273 (1980).
4. V. Yakhot, preprint.

NEGATIVE CALCIUM IONS ARE STABLE— IN SPITE OF THE CONVENTIONAL WISDOM

Most atomic species—roughly 80% of the periodic table—can form stable negative ions in their gaseous phase. The most notable and easily understood exceptions are the noble gas atoms, the symmetry of whose completely filled electron shells offers no handhold for a would-be extra tenant. The alkaline earths—calcium, strontium, barium and their group IIA cousins—are similarly inhospitable, or so the standard textbooks assure us. Two back-to-back reports in a recent issue of *Physical Review Letters* have now disabused us of this venerable assurance.

David Pegg and coworkers at the University of Tennessee and Oak Ridge have provided the surprising experimental demonstration that calcium does, after all, form stable negative ions.[1] At the same time, Vanderbilt University theorist Charlotte Froese Fischer and collaborators at the University of Toronto report[2] the results of an ambitious multiconfigurational Hartree–Fock calculation which tells us that a neutral calcium atom ought indeed to bind an extra electron in a surprising valence configuration, with a fragile binding energy of about 0.045 electron volts—in striking agreement with what Pegg and company see.

Holding an extra electron

A neutral atom does not, of course, exert any net Coulomb force on an external electron. But if the extra electron is close enough to penetrate partially into the atom's electron cloud, it will experience a net attraction. The deeper the visitor penetrates, the less effectively does the cloud shield the positive charge of the nucleus. Furthermore, the repulsive force of the extra electron tends to polarize the atomic electron distribution to some degree, subjecting the intruder to higher-multipole attractive forces.

In both cases the electrostatic attraction of the extra electron to the atom falls off significantly faster with distance than does a simple Coulomb force. Therefore it is easy to see that in the noble gas case, where the Pauli exclusion principle restricts the extra electron to the next principal or orbital-angular-momentum quantum number beyond the filled shells, the attraction is too weak to permit a stable negative ion. Similar arguments are traditionally invoked for calcium and the other alkaline earths, whose two valence electrons form a closed spin-singlet S subshell. By contrast, sodium, potassium and the other alkali metals, with only a single unpaired valence electron, all do form weakly bound stable negative ions. The most strongly bound negative ions, of course, are to be found among the halogens—fluorine, chlorine and the others—whose outermost shells are just one shy of full occupancy. Note that we are speaking here of single atoms in the gas phase. Negatively charged ions in solution or embedded in crystals gain additional stability from their surroundings.

As we now know, qualitative arguments and traditional Hartree–Fock calculations are grossly inadequate for predicting whether the alkaline earths form stable negative ions. The binding energy of the extra electron in Ca^- is less than 10^{-5} of the total binding energy of the 20 electrons of Ca^0. The negative ion is stable if its total binding energy is greater (more negative) than that of the neutral atom's ground state. One calls the difference between these total energies, $E^0 - E^-$, the "electron affinity" of the neutral atom. When it is positive it is essentially the binding energy of one extra electron. When it is negative, the extra electron will sooner or later "autodetach," the ion reverting to the neutral atomic state. Calculating the sign and magnitude of the electron affinity "is like trying to weigh a ship's captain by weighing the ship before and after he comes aboard," suggests Pegg.

Despite the fact that their treatment of interelectron correlations is rudimentary, the usual "independent-particle model" Hartree–Fock computer calculations can generally reproduce the total energies of atomic electron configurations to within a percent of the experimental value. That's adequate for many purposes, but not for calculating electron affinities, which are typically much smaller than the ionization potentials of the corresponding neutral atoms. One is trying to get a tiny difference between two large numbers, having ignored correlation effects much larger than this difference. Froese Fischer, by undertaking an extensive treatment of these previously slighted correlation effects on a Cray supercomputer, becomes the first to get the sign of the calcium electron affinity right.

The experimental problem is equally delicate. With the extra electron so loosely held, most "stable" negative ions are quite fragile in the gas phase. They are hard to make and easy to destroy. It is difficult to accumulate them in sufficient numbers to do conventional optical spectroscopy. One usually resorts to photoelectron spectroscopy with crossed ion and laser beams. Stray voltages, high backgrounds and low photodetachment cross sections make it very difficult to measure the low-energy photoelectron spectrum accurately.

Pegg, his University of Tennessee student Jeffrey Thompson, and their Oak Ridge collaborators Robert Compton, Gerald Alton and Thomas Kvale had originally built their apparatus at Oak Ridge to look for autodetached electrons from what they assumed were *metastable* Ca^- ions. Failing to find the expected signal of the spontaneous decay of Ca^- to Ca^0, and having told Froese Fischer of this puzzling failure, they went on to show that their negative calcium ions were, in fact, stable. The extra electron of a metastable negative ion comes off by itself; one has only to wait. To study the binding of a stable negative ion, on the other hand, the Tennessee–Oak Ridge group had to add a laser to provide the energy needed to photodetach the extra electron.

Metastable negative ions

The ground state valence-electron configuration for all the group IIA elements is written ns^2 (^{1}S), where n is the principal quantum number for the valence shell, s^2 tells us it contains two s-wave electrons, and (^{1}S) gives the total orbital and spin angular